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DEVELOPMENT OF PUMPS FOR PULSE COMPRESSION LASER APPLICATIONS.(U)
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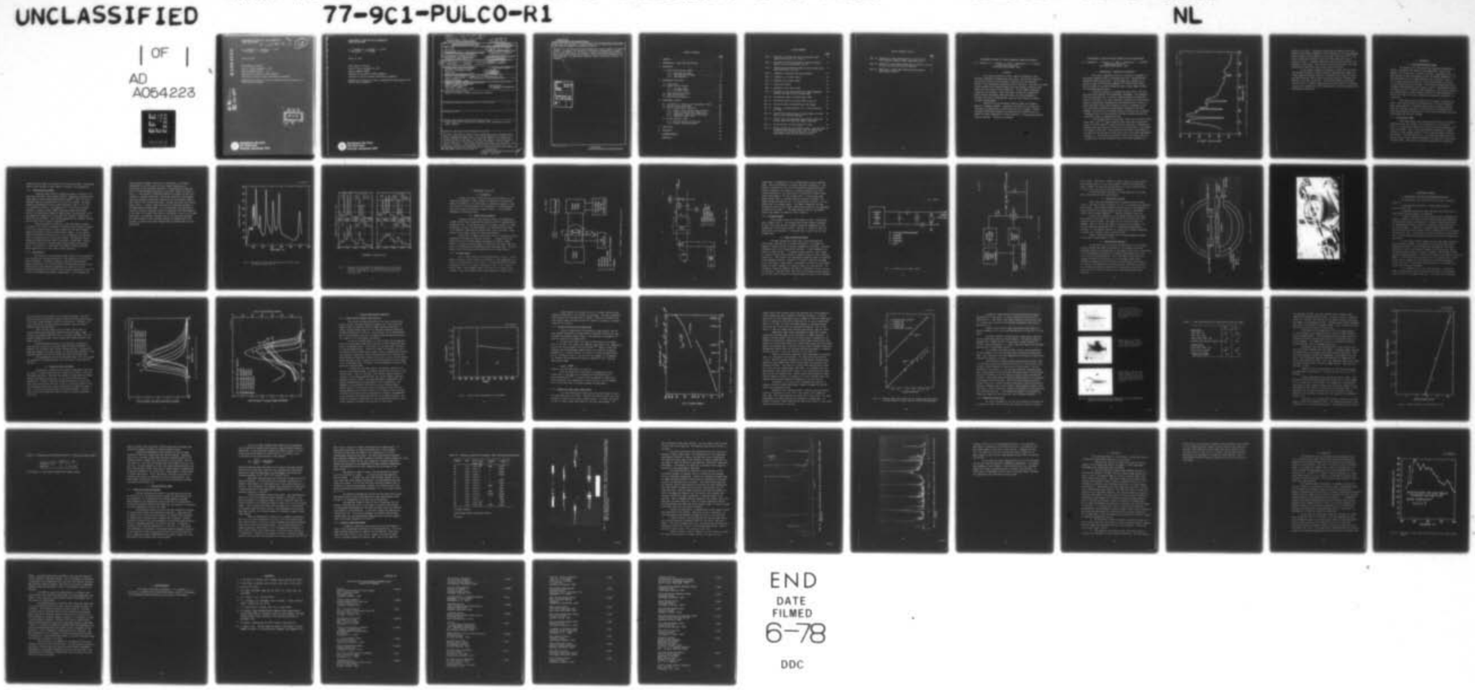

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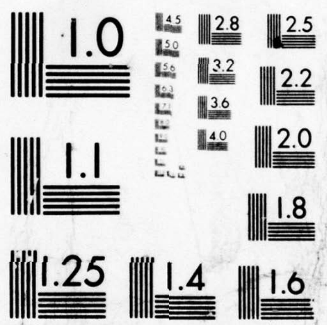

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DEVELOPMENT OF PUMPS FOR PULSE COMPRESSION
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N. T. Melamed, R. J. Zollweg, J. L. Pack,
T. Henningsen, J. T. Veligdan

March 10, 1978

Final Report for Period

June 15, 1976 to December 15, 1977

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Westinghouse R&D Center
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This report describes an effort aimed at developing a more efficient flash-lamp for pumping blue laser dyes. The work was supported by the Navy as part of their program to develop laser sources which operate in the blue-green transmission band of sea water. The effort was based on the possible use of mercury lamps of the high pressure capillary type (AH6), with and without spectral additives. A wide range of operating conditions were explored. Those found most suitable for pumping Rh6G gave no better performance than xenon

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lamps, and tests designed to evaluate their use for pumping blue laser dyes also showed no evidence of improved performance.

Additives for the spectrally enhanced lamps were chosen by means of a computer program. In general, the spectra of the additives conformed in a general way to those predicted by the program. However, operating difficulties primarily arising from lamp instabilities, precluded the useful application of these additives. An alternative approach, based on molecular radiators, is proposed.

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Westinghouse R&D Center
Pittsburgh, Pennsylvania 15235

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Pittsburgh, Pennsylvania 15235

1. INTRODUCTION: OBJECTIVES AND APPROACH

This report summarizes an effort to develop a flashlamp for efficiently pumping a blue or blue-green laser dye. The motivations behind the effort were several. The principle one, however, was to provide a means for effectively pumping a Pr^{3+} doped crystal laser, which, because of its narrow absorption bands is best pumped by means of a blue emitting dye laser.

The major difficulty in pumping blue laser dyes is the very poor efficiency obtained with conventional flashlamps. The reasons for this poor efficiency lie in the poor overlap between the absorption bands of these dyes in the near ultraviolet with the very broad spectral output of conventional flashlamps, and the relatively low uv output of xenon flashlamps, whose spectra approach those of a blackbody at the very high current densities and peak powers required for dye laser pumping. The purpose of the present project, therefore, was to develop a flashlamp which is a more selective radiator and better suited than conventional lamps for pumping blue dyes.

The starting point for the present effort was a series of papers by Dal Pozzo, et al[(1-3)] which predicted that pulsed mercury capillary lamps of the AH-6 family[(4)] should prove more efficient at pumping blue coumarin laser dyes because of their better spectral match to the absorption bands of these dyes. This prediction was based on measurements of the spectra of pulsed mercury lamps rather than on dye laser measurements (see Fig. 1). The authors were unable to obtain any laser action with a coumarin dye, and only threshold laser action with extremely poor

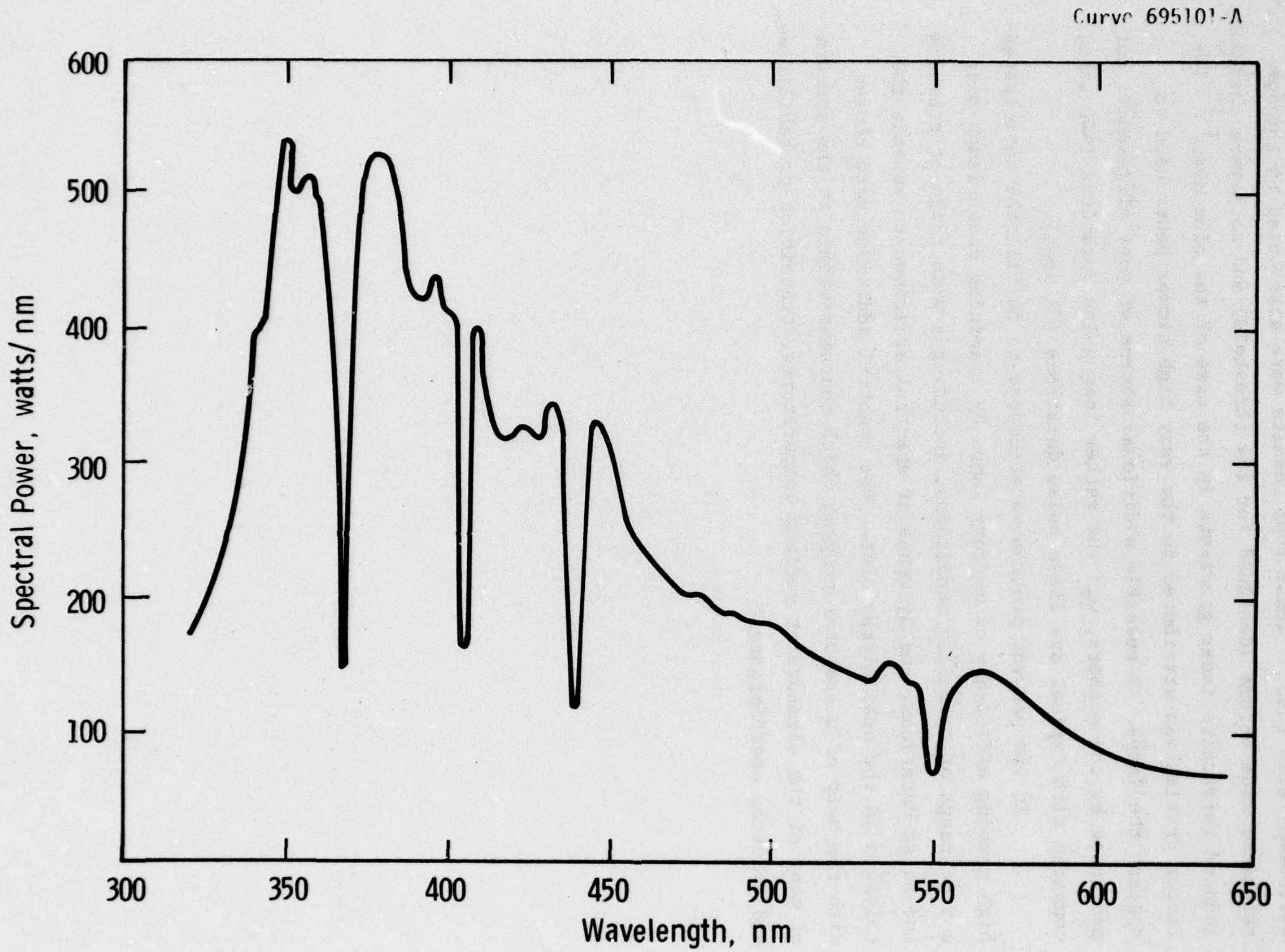

Fig. 1. Spectrum of a pulsed AH-6 mercury capillary lamp (after Dal Pozzo, et al., Ref. 1).

efficiency with Rh6G. These poor results were attributed to the low resonator mode volume (designed for low threshold) and to severe thermally induced refractive index gradients in the case of the blue dye.[3] The latter problem was attributed to the very high simmer power used to operate the lamps. A possible additional source of poor efficiency, not mentioned by the authors, was the rather long pulse durations ($\sim$40 μsec) compared with typical dye laser pulse durations ($\lesssim$1 μsec).

In the present program we attempted to fulfill the anticipated high pumping efficiency of mercury lamps by examining these lamps over a wide range of operating conditions, including a wide range of pressure and pulse durations, and by means of spectral additives to enhance the emission in the near ultraviolet. The spectral additives were chosen with the help of a computer program which contains within it the spectra of most of the elements at various temperatures, transition probabilities, and emission coefficients.

2. BACKGROUND

2.1 High Pressure Mercury Lamps

The operation of a high pressure Hg lamp may be understood as follows: Initially, the lamp is at room temperature. Most of the mercury is in liquid form. As power is applied, a discharge occurs at a vapor pressure of Hg corresponding to Hg at the ambient temperature. The heat developed by the arc causes additional Hg to vaporize. This, in turn, causes the mercury pressure to rise, which produces an increase in the voltage drop across the lamp. The increased voltage drop results in higher lamp dissipation which results in more of the Hg being vaporized and causes a further increase in lamp dissipation. This process continues until equilibrium is reached between the amount of power dissipated in the arc and that lost to the surroundings by radiation, conduction, and convection.

High pressure mercury lamps are of two types: dosed and overdosed. Dosed lamps contain just enough mercury to be fully vaporized under normal operation. Overdosed lamps contain an excess of mercury under all operating conditions. The distinction is an important one, there being significant differences in the electrical and spectral properties between the two types of lamps.

2.1.1 Dosed Mercury Lamps

Dosed lamps contain just enough mercury so that, at the normal operating current and voltage, all of the mercury is in the form of vapor. As in all mercury lamps, the pressure rises until operating conditions are reached. Once at the normal operating conditions, a further increase in power loading results in only a relatively small increase in gas pressure. The lamp spectrum, i.e., the relative intensities of the various spectral lines, remains approximately the same over a wide range of input powers. Because of this relative insensitivity to operating

conditions, most lamps in use today are of the dosed variety. The maximum mercury vapor pressure of dosed lamps is typically a few atmospheres.

2.1.2 Overdosed Mercury Lamps

Overdosed lamps contain a substantial excess of mercury at all operating conditions. The prototype lamp for commercially available overdosed lamps is the AH6 high pressure mercury lamp.[4] This is a quartz capillary lamp with a 2 mm bore and a 2.5 cm (1") arc length. The lamps normally are operated in a water cooled housing at 850 V, 1.2 A, and at a pressure of 100 atm. Arc lengths of 2", 3" and 4" are currently available. Except for their higher operating voltages and radiant outputs, (the operating voltage and radiant power is proportional to the arc length) the properties of these lamps are the same. Narrower bore lamps are also available. Their spectral properties are somewhat modified but are still close to those of the AH6.[5]

Since the quantity of mercury in over-dosed lamps is greatly in excess of that which is vaporized, the mercury vapor pressure continues to be a very strong function of power loading over the entire range of operating conditions within the high pressure regime. (A low pressure regime is discussed in Section 4.3.1.) As a consequence, the lamp spectrum is a strong function of the power loading. The ballasted current-voltage characteristics also differ from those of dosed lamps. Because the mercury pressure of overdosed lamps varies strongly with applied power, it is possible to investigate the behavior of these lamp under pulsed conditions over a wide range of vapor densities.

2.1.3 Lamp Spectrum

The spectrum of a high pressure lamp consists of two components--line radiation and continuous radiation. Line radiation is due to bound-bound transitions. Continuous radiation arises from three sources: molecular radiation, due to mercury dimers, recombination radiation (free-bound) due to a free electron recombining with a mercury ion, and bremsstrahlung radiation (free-free) due to electrons scattered by atoms.[6] At very low pressures line emission predominates. As the pressure and

power loading is raised, there occurs a broadening of the lines, a suppression of the 185 nm and 253.7 nm resonant radiation, and an increase in the continuous radiation. The spectrum of an AH6 lamp with a 1" long discharge operating at rated power (1000 W) is shown in Fig. 2. A fair amount of background radiation is present and the 253.7 radiation is largely suppressed. The energy contained in the resonant radiation reappears in molecular bands and in the non-resonant lines at 366.3, 404.8, 435.6 nm and other lines. Another spectrum of this lamp is shown in Fig. 3. Figure 3 shows the effect of increasing the power to the lamp beyond the rated power. It is evident that the proportion of continuous radiation increases rapidly in comparison with line radiation as the power is increased, even under cw conditions, so that, at 2.5 times rates power, the spectrum is largely that of the continuous radiation. The rapid increase in the continuous radiation is largely due to the increase in pressure with power loading which is characteristic of over-dosed lamps.

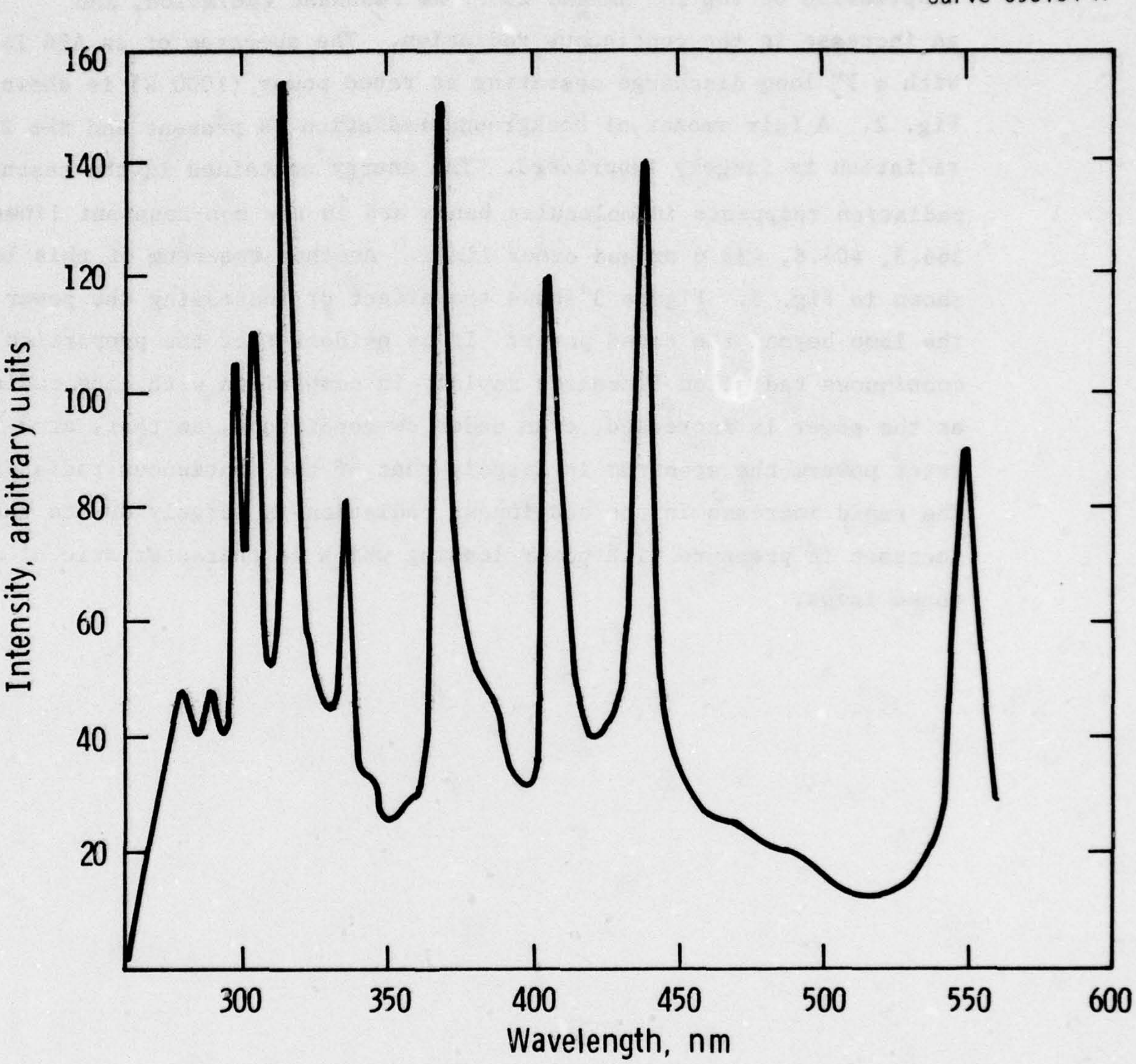

Fig. 2. Spectrum of an AH-6 high pressure mercury capillary lamp at 100 atm (after Stahl, Ref. 5).

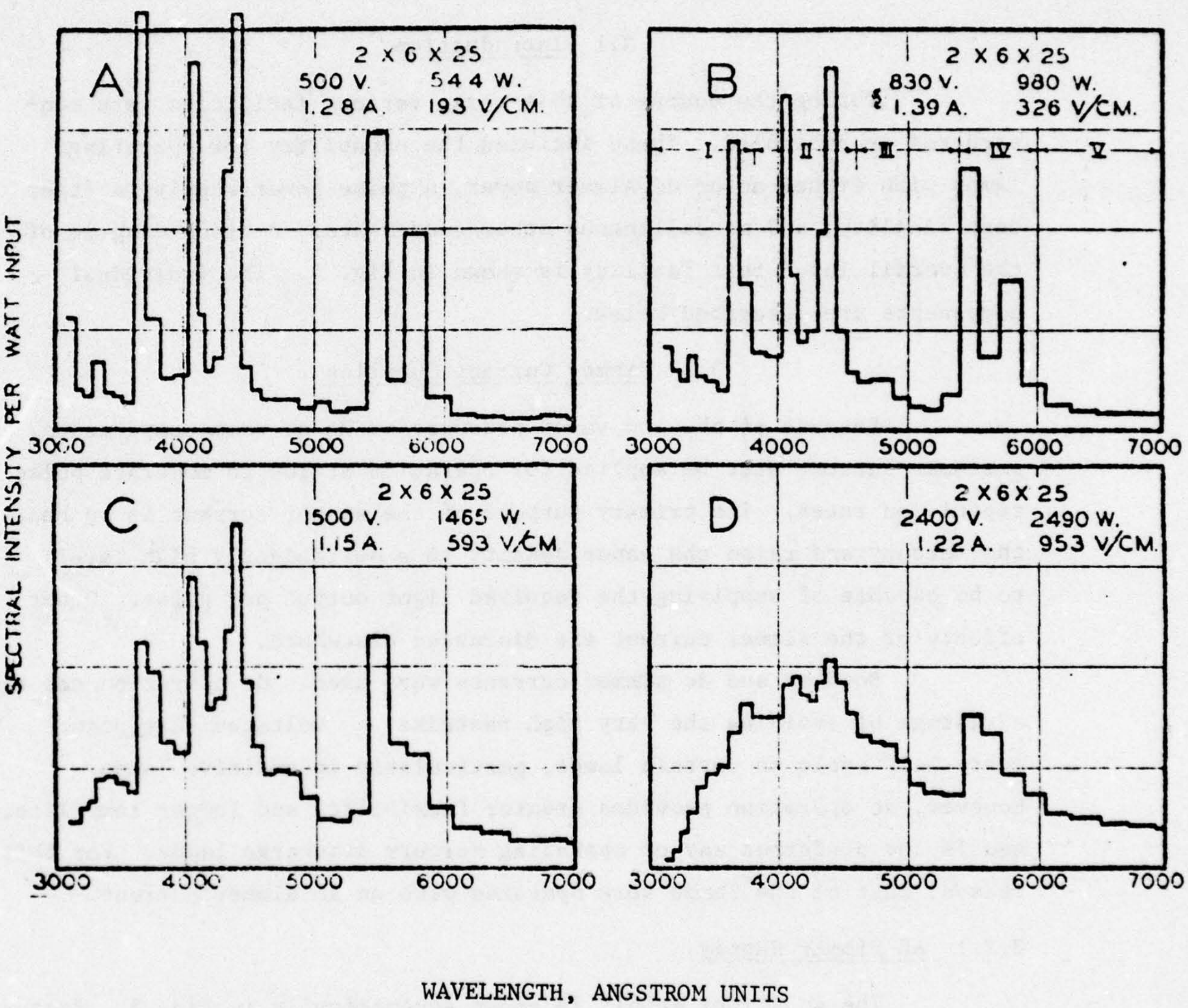

Fig. 3. Spectra of an AH-6 capillary lamp operated at various power loadings and pressures: A, 200 V/cm, 33 atm; B, 332 V/cm, 77 atm; C, 600 V/cm, 167 atm; D, 960 V/cm, 287 atm (after Noel, Ref. 4a).

3. EXPERIMENTAL FACILITIES

3.1 Introduction

During the course of this work, various facilities were constructed or assembled. These included the capability for operating lamps with either ac or dc simmer power, a pulse power supply, a laser test facility, and miscellaneous other components. A block diagram of the overall laser test facility is shown in Fig. 4. The individual components are described below.

3.2 Simmer Current Supplies

Because of the low vapor pressure of Hg at room temperature, a simmer current must be applied for operation at low to moderate pulse repetition rates. The primary purpose of the simmer current is to heat the mercury and raise the vapor density to a sufficiently high level to be capable of supplying the required light output per pulse. Other effects of the simmer current are discussed elsewhere.

Both ac and dc simmer currents were used. dc operation has the advantage of avoiding the very high restrike voltages that occur every half cycle in certain lamps, particularly in additive lamps. However, ac operation provides greater flexibility and longer lamp life, and is the preferred way of operating mercury discharge lamps. For this reason, most of the lamps were operated with an ac simmer current.

3.2.1 AC Simmer Supply

The ac simmer supply is shown schematically in Fig. 5. Because additive lamps are often difficult to stabilize, a wide ballasting capability was incorporated into the supply. This had the additional benefit of making possible a wide range of operating conditions for standard mercury lamps. Both variable inductive and resistive ballasting were included. The main component of the power supply was a 3000 V saturable

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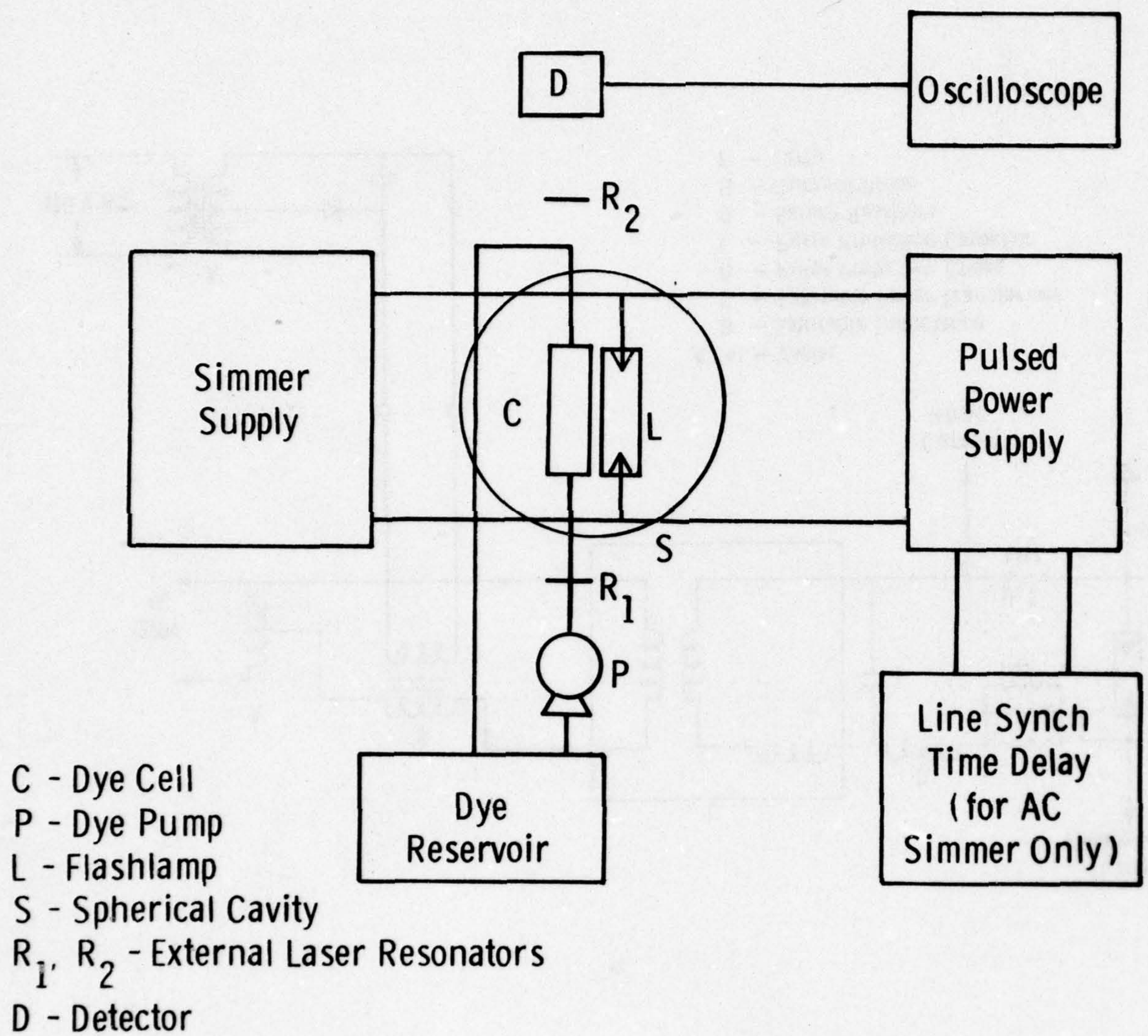

Fig. 4. Schematic of lamp/dye laser test facility.

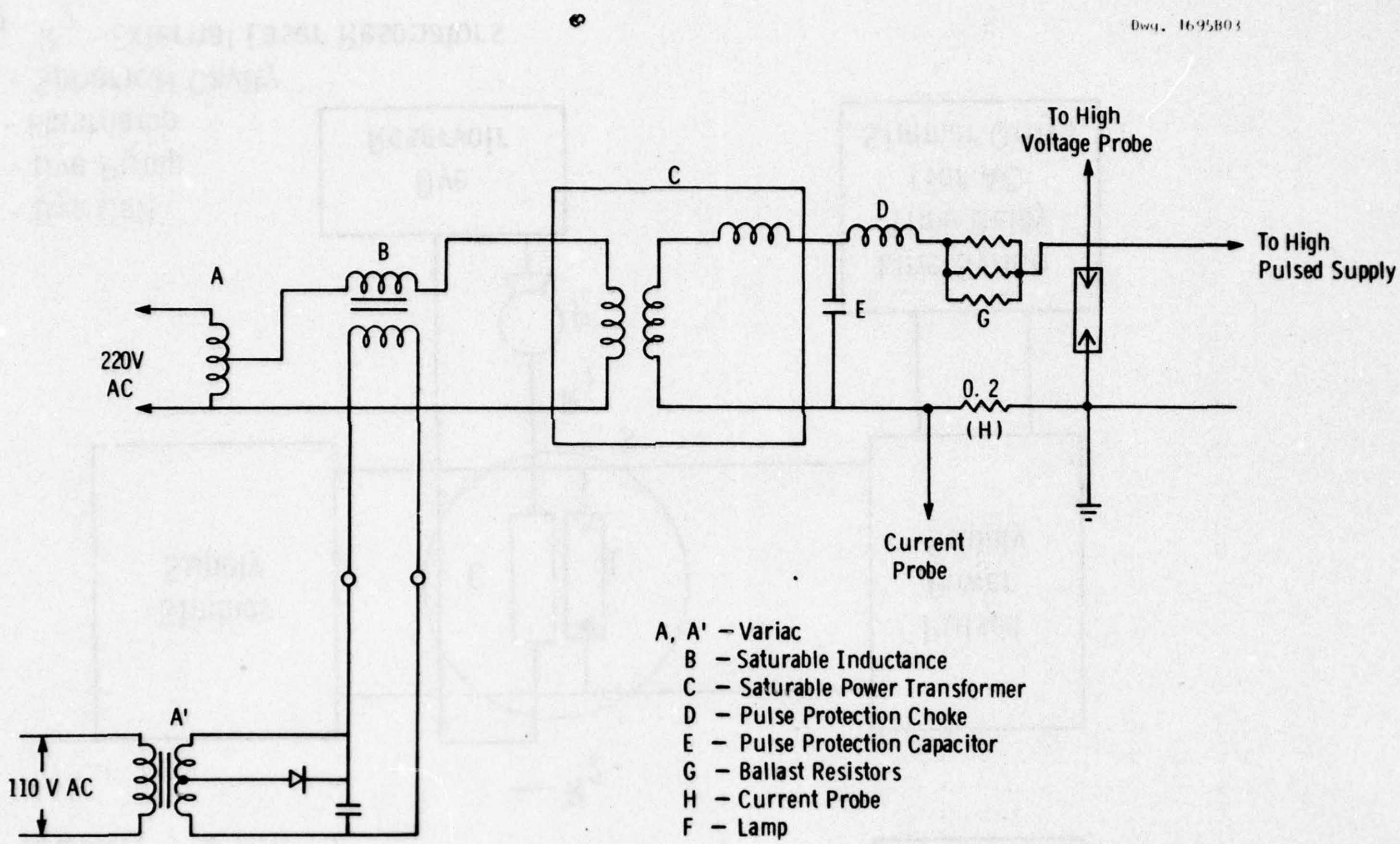

Fig. 5. Schematic of ac simmer supply.

transformer, C, designed for use with high pressure mercury capillary lamps. This was preceded by a variable saturable inductance, B. The degree of saturation of the core of the variable inductance could be varied by applying a dc current to a secondary winding on the core. A small inductance, D, and capacitance, E, protected the 3000 V transformer from damage by the high voltage pulse. A high voltage high power resistor bank, G, provided resistive ballast, if needed. The simmer current was monitored by a 0.2 ohm precision resistor, H, in the ground side of the simmer supply. The simmer voltage and the pulse voltage were monitored by a high voltage probe attached to the ungrounded electrode of the lamp, F . The simmer voltage could be continuously varied from 0-3000 V by means of a 220 V variac, A.

3.2.2 DC Simmer Supply

The dc simmer source is a much simpler circuit in principle, and is shown in Fig. 6. The variable high voltage supply, A, consisted of one or more line operated variable dc supplies, depending on need. Voltages up to 30 kV were available. Resistor B served to limit the lamp current, and capacitor C protected the dc supply from the high voltage pulse.

3.3 High Voltage Pulse Supply

The high voltage pulse source was an existing home-built power supply, modified for the present application. The supply was capable of either internal or external triggering, with either a variable repetition rate or with single pulse operation. A block diagram of the circuit is shown in Fig. 7. An energy storage capacitor C was charged to a preset voltage from a variable dc supply through a current limiting resistor R-1. The capacitor was discharged through the lamp by triggering a 5C22 hydrogen thyratron. Resistor R-2 insured a capacitor discharge loop in the event of lamp failure. Inductance L was used only as required either to prevent ringing or to produce a pulse of a desired width. The voltage to charge the capacitor was kept to a maximum of 14 KV to protect the thyratron. The most widely used value of the energy storage capacitor was 0.1 μfd, which resulted in a maximum energy per pulse to the flashlamp

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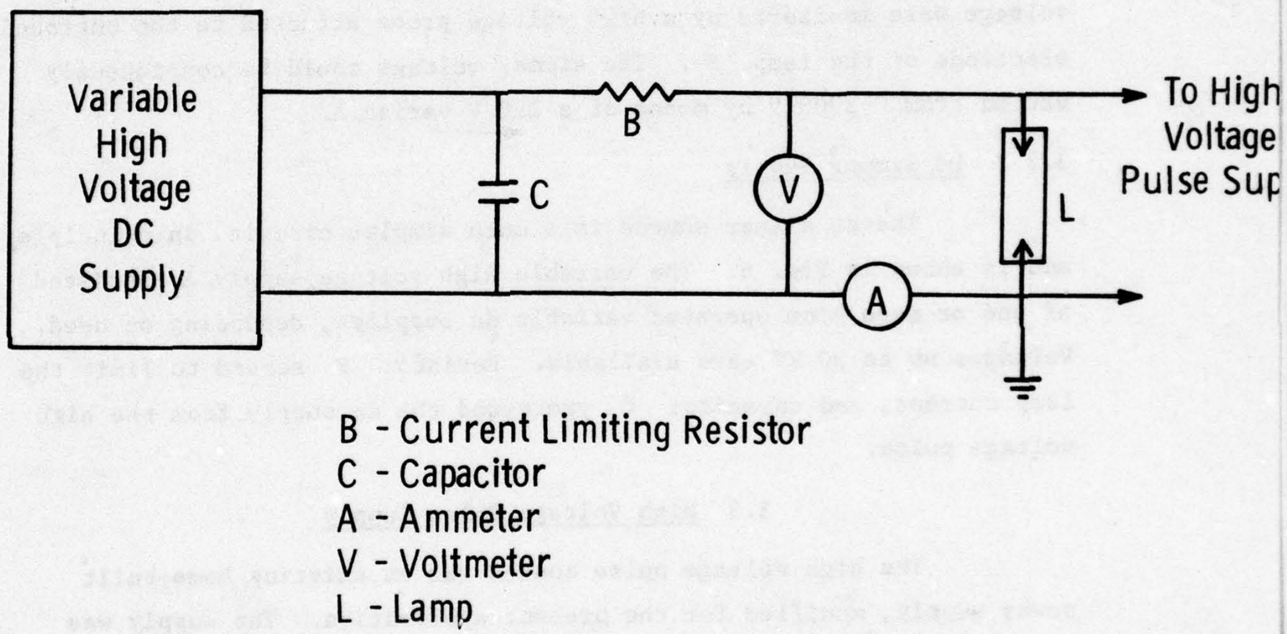

Fig. 6. Schematic of dc simmer supply.

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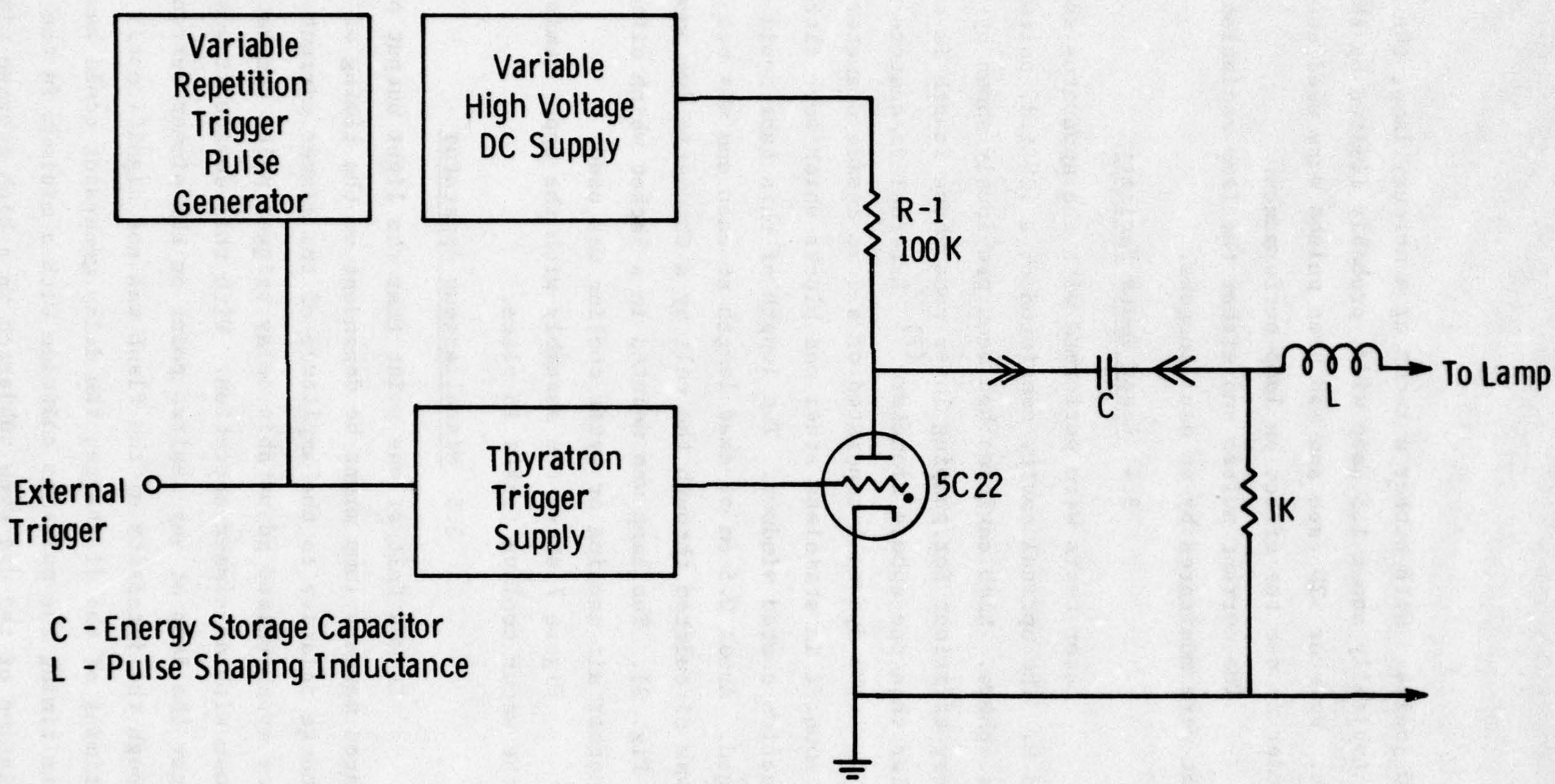

Fig. 7. Pulse power supply.

of 10 joules. With either a xenon or a mercury lamp, the current pulses were typically about 1.5 μsec wide, probably limited by the lamp impedance. However ∿20 μsec and ∿50 μsec pulses were used at various times in order to see the effect on lamp performance.

The current pulses and either the lamp radiation or the laser output were monitored by an oscilloscope.

3.4 Laser Tests Facility

Laser tests were performed with the apparatus shown in Figs. 8 and 9. The optical cavity consisted of a 12" i.d. polished and aluminized glass sphere. Such cavities had been previously shown by Westinghouse to be very efficient for pumping laser rods whose length is considerably smaller than the sphere diameter.[7] External resonators were used.

The dye cell consisted of a 2 mm inside diameter fused silica tube mounted in stainless steel end blocks which were fitted with antireflection coated windows. The length of this tube could easily be changed. About 0.5 cm of dead length at each end was not pumped. The dye was circulated through the cell by a Chromatix dye pump module (see Fig. 2). The lamp was mounted in a jacket which differed depending on whether air cooling or water cooling was used.

Figure 7 shows the assembly with the upper hemisphere removed and the water cooling jacket in place.

3.5 Miscellaneous Apparatus

It was felt at one point that the light output of an ac simmer operated mercury lamp might be dependent on the timing of the pulsed discharge relative to the amplitude of the simmer current wave. Accordingly, a line synchronized adjustable delay trigger pulse generator was built for use with ac simmer operation. With this device it was possible to trigger the lamp at any desired point on the simmer current wave. Although the intensity of the flash was not significantly influenced by the timing of the discharge, the delay generator could have a potential use in timing the pulse to coincide with a minimum in the triplet state population of the dye when subjected to a high average simmer power radiation.

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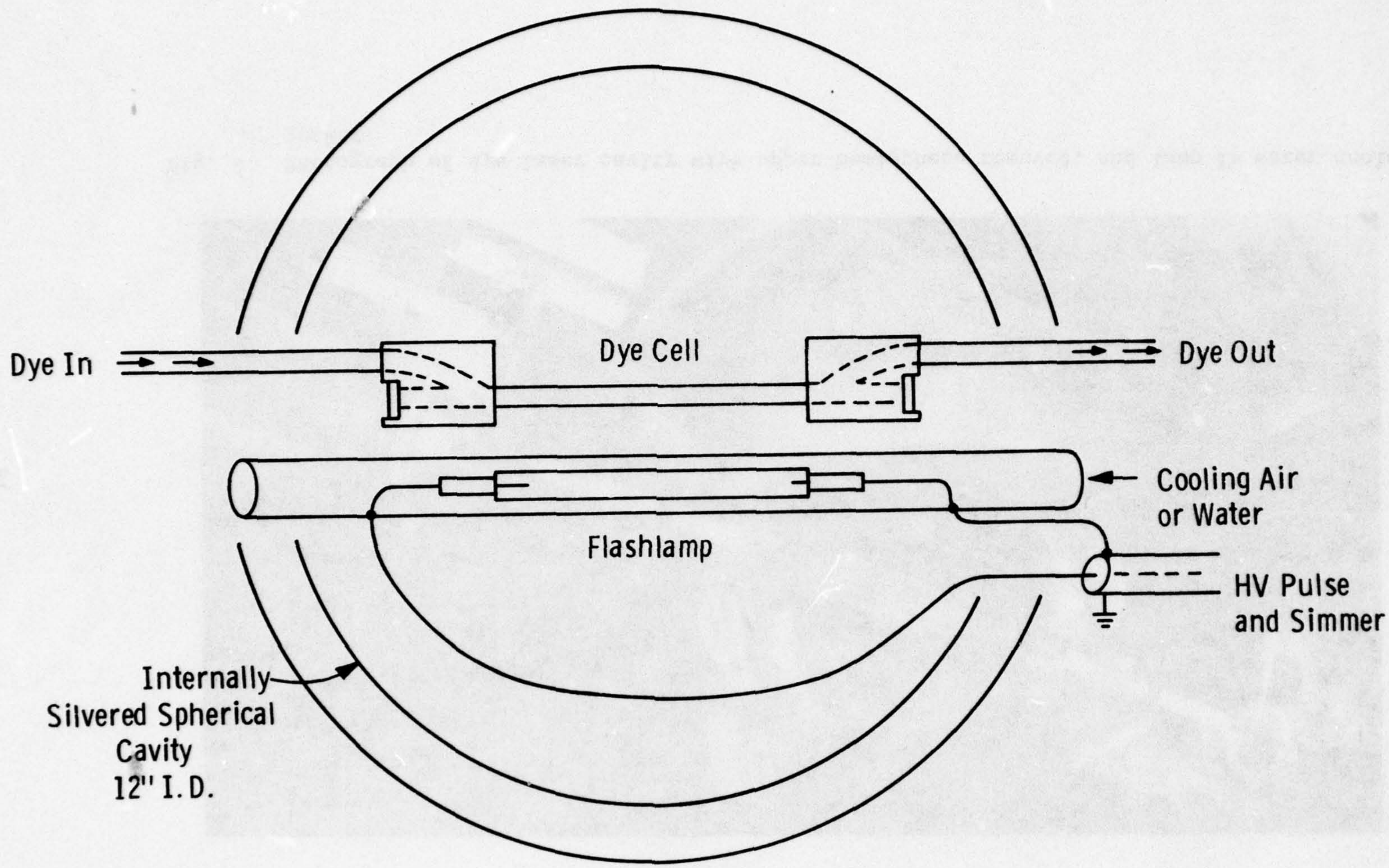

Fig. 8. Schematic of dye laser cavity.

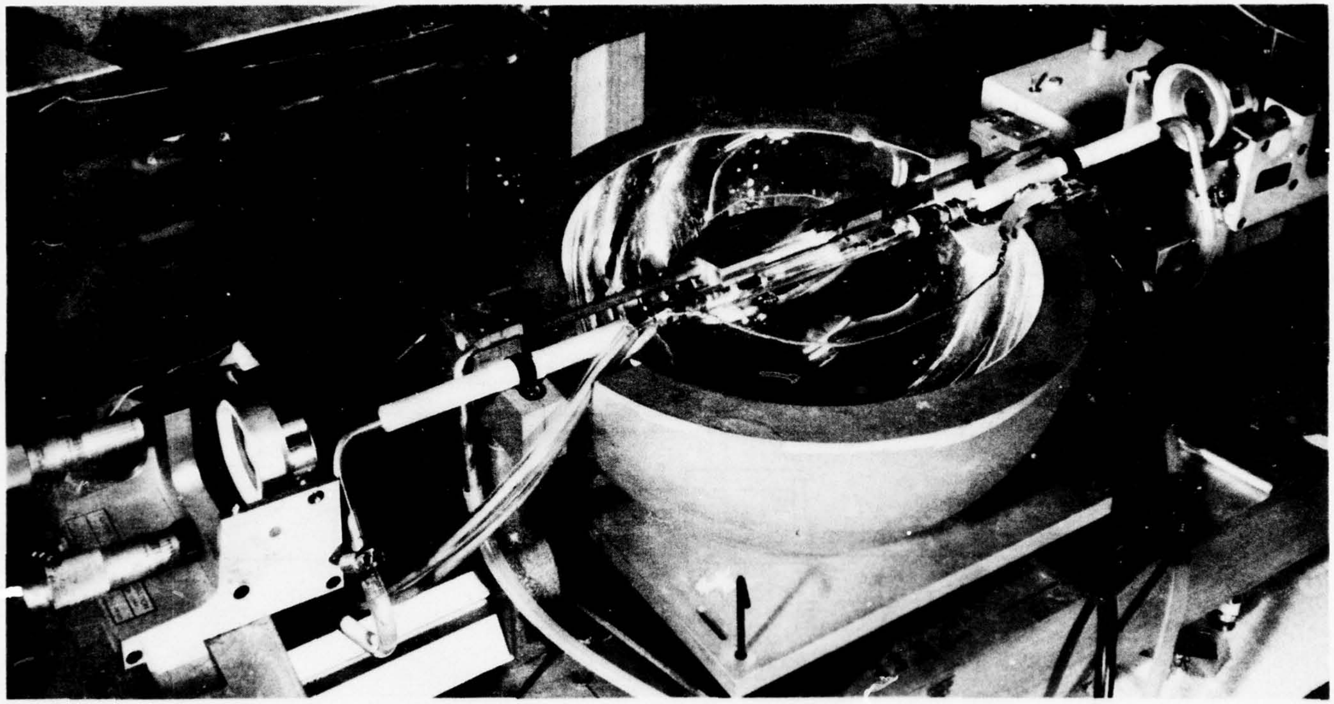

Fig. 9. Photograph of dye laser cavity with upper hemisphere removed, and lamp in water cooled jacket.

4. EXPERIMENTAL RESULTS

4.1 Introduction: Outline of the Experimental Effort

The experimental work initially took the form of a parallel effort to

a) characterize the behavior of high pressure mercury capillary lamps, and

b) determine which metal vapor additives were best suited for enhancing the desired region of the spectrum.

In order to establish in which spectral region enhancement was desired, a number of blue laser dyes were obtained and their absorption and excitation spectra were measured. A "typical" excitation spectrum was established and used as a model to determine suitable additives for the additive lamps. In addition, a detector-filter combination was constructed whose spectral response closely resembled that of the "typical" dye excitation curve. This detector was used to determine the effectiveness of various lamps and different operating conditions for pumping blue laser dyes.

A total of more than 40 lamps, both with and without additives were tested and consumed, often because of lamp failure under severe operating conditions. Tests included those performed in a dye laser apparatus, as well as the spectral response tests noted above. None of the additive lamps reached the stage of testing in the dye laser, mainly because of lamp instabilities. In the final stage of this work, an effort was made to a) define the best conditions for use of a mercury lamp to pump a laser dye; b) compare the effectiveness of a pulsed high pressure mercury lamp with a conventional xenon flashlamp, and c) determine the limitations of mercury flashlamps.

It is important to note that the properties of the mercury lamps, including their behavior under pulsed operation are determined almost entirely by the simmer conditions. The simmer power determines

the vapor density of the mercury at low repetition rates. The vapor density does not significantly change during an individual pulse because of the high thermal inertia involved in vaporization. Estimates of the vapor pressure and the number density of mercury atoms were made at different operating conditions.

Apart from the central role the simmer current plays in establishing the operating vapor pressure of mercury lamps, simmer currents are generally desirable even in rare gas flashlamps. They make possible higher peak power, shorter pulses, more stable operation, and longer lamp life.

Since operating stability was an important consideration, some attention was paid to the usable range of simmer conditions. In addition, air cooling as well as water cooling was used. Although the lamps are designed to be water cooled, air cooling provided access to some operating regions that were not stable when water cooling was used.

The experimental results are described below. They are arranged for continuity of presentation and not in the order in which they were obtained.

4.2 Spectra of Blue Laser Dyes

Fluorescence, absorption and excitation measurements were made on a group of coumarin dyes which were judged to be typical of blue and blue-green laser dyes. More than one solvent was used where the solvent strongly influenced the spectrum. All dyes were obtained from the Exciton Corp. Figure 10 shows the fluorescence spectra of these dyes. Their excitation spectra are shown in Fig. 11; excitation spectra rather than absorption spectra are shown because they are judged more significant in defining the optimum region for flashlamp pumping. The band pass of the filter combination which was used to evaluate flashlamps is also shown.

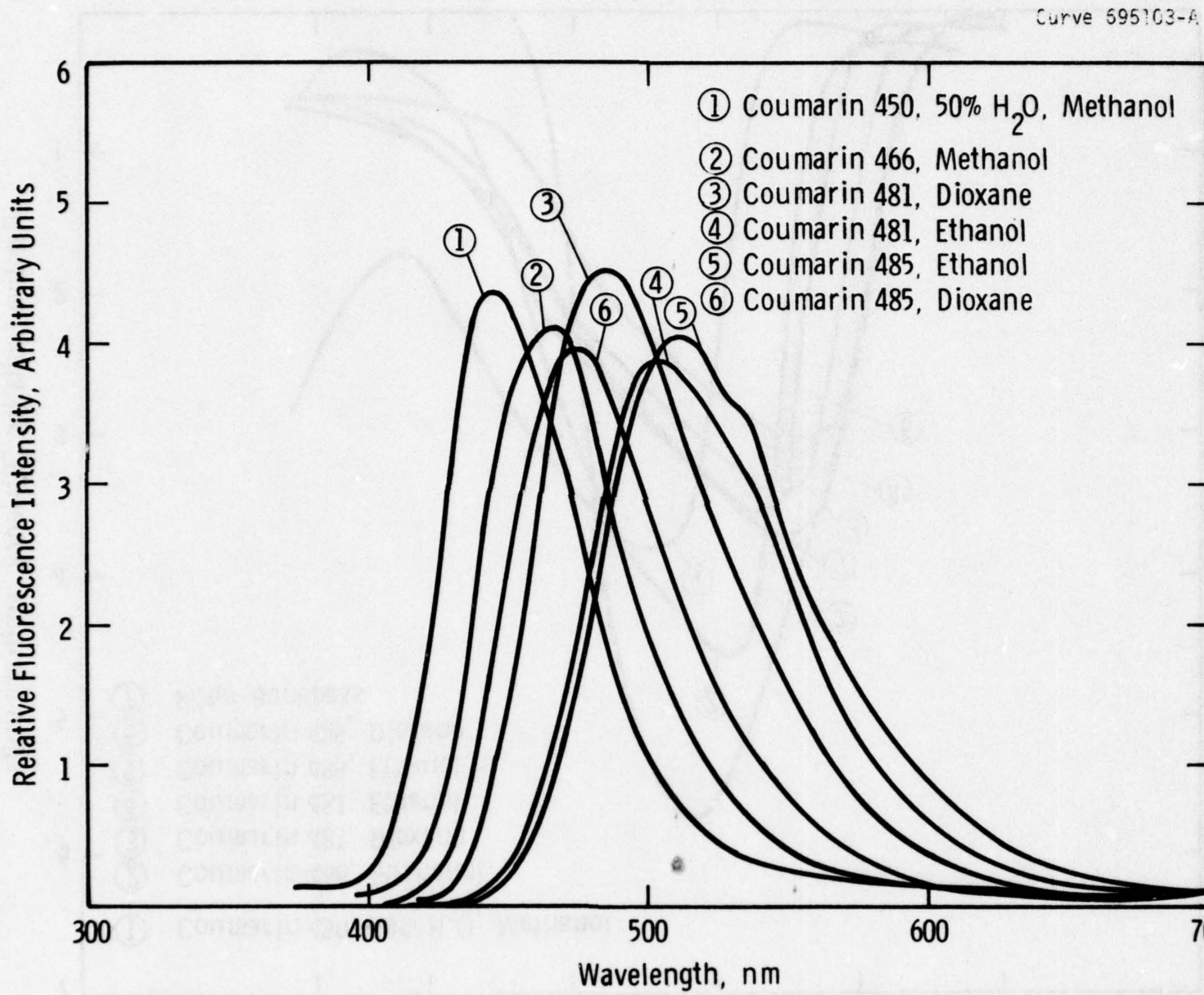

Fig. 10. Fluorescence bands of selected laser dyes.

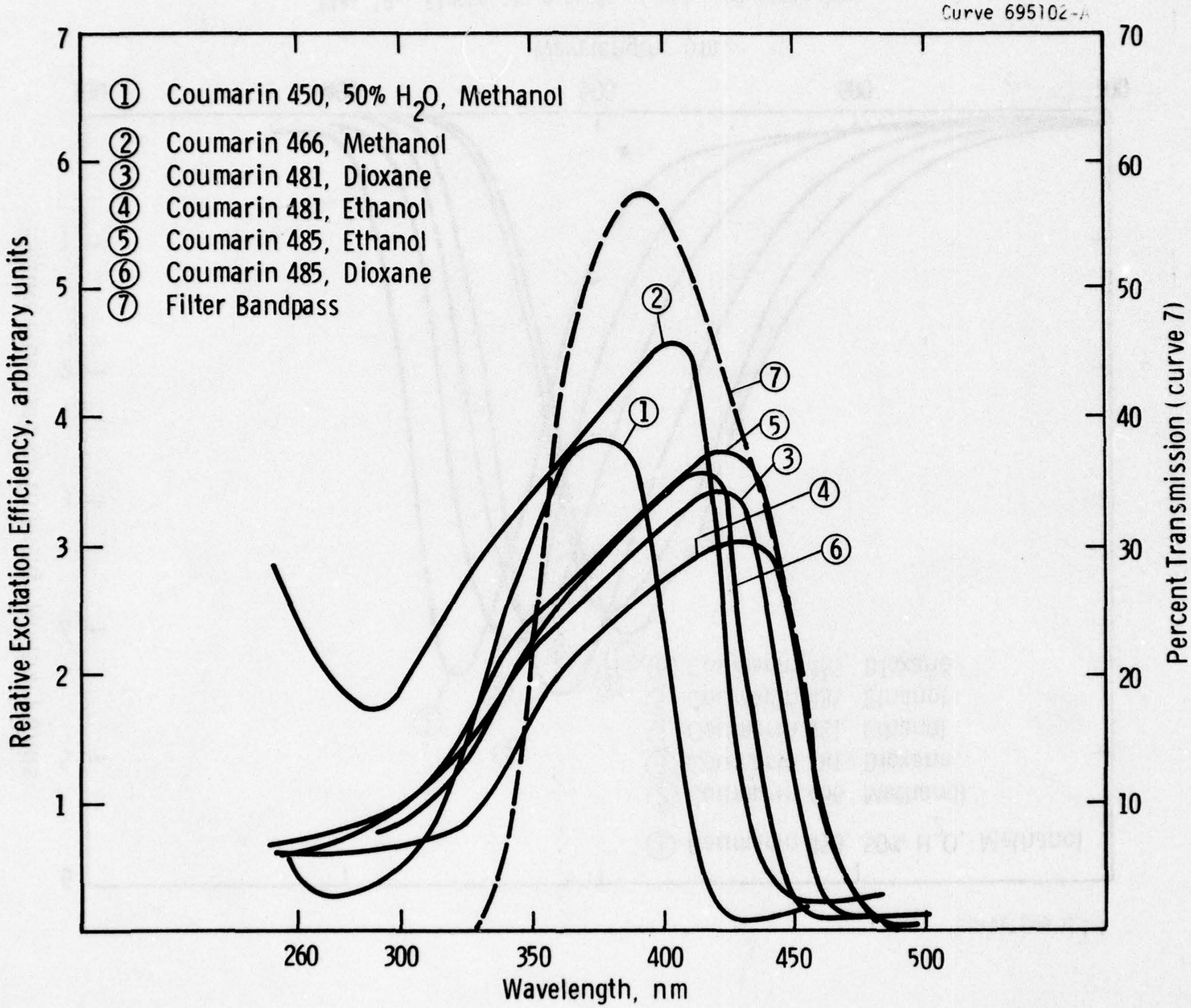

Fig. 11 Excitation bands for the dyes shown in Fig. 10

4.3 Mercury Lamps Without Additives

4.3.1 Simmer Current-Voltage Characteristics

The current-voltage characteristics of the ac operated high pressure mercury capillary lamps were measured over their operating range in order to determine suitable simmer operating conditions. A typical current-voltage trace for a 2" lamp with inductive ballasting is shown schematically in Fig. 12. Three regions can be distinguished. Two of these, labeled I and III, are stable operating regions separated by a region, II, of considerable instability.

In the low voltage region, I, ($\sim$25-35 V/cm), the voltage drop across the lamp remains essentially constant, actually dropping slightly with an increasing current. The current, on the other hand, continues to rise with applied power. Within this region the behavior is that of a conventional low pressure mercury lamp in which increased power involves the creation of additional current carrying electrons by collision with atoms causing the resistance to drop, the current to rise, and the voltage across the lamp to drop. Evidently the power dissipation in this regime is not sufficiently great to produce a strong dependence of mercury vapor pressure on applied power.

In region III the lamp exhibits the behavior typical of the high pressure operating mode. The region extends from $\sim$150 V/cm to $\sim$400 V/cm and above. Here, an increase in the applied voltage produces an increase in the voltage gradient while the current remains essentially constant or drops a little. The lamp resistance, as measured by the average characteristics, is proportional to the voltage drop. The behavior is the converse of that seen in the low pressure region. Within region III, the mercury pressure increases uniformly with the voltage gradient, so that the increase in the number of free electrons produced by the higher field is compensated or slightly overcompensated by the decrease in the mean free path due to the higher pressure. The positive resistance characteristic of the lamp-ballast combination makes stable operation relatively easy over most of this range.

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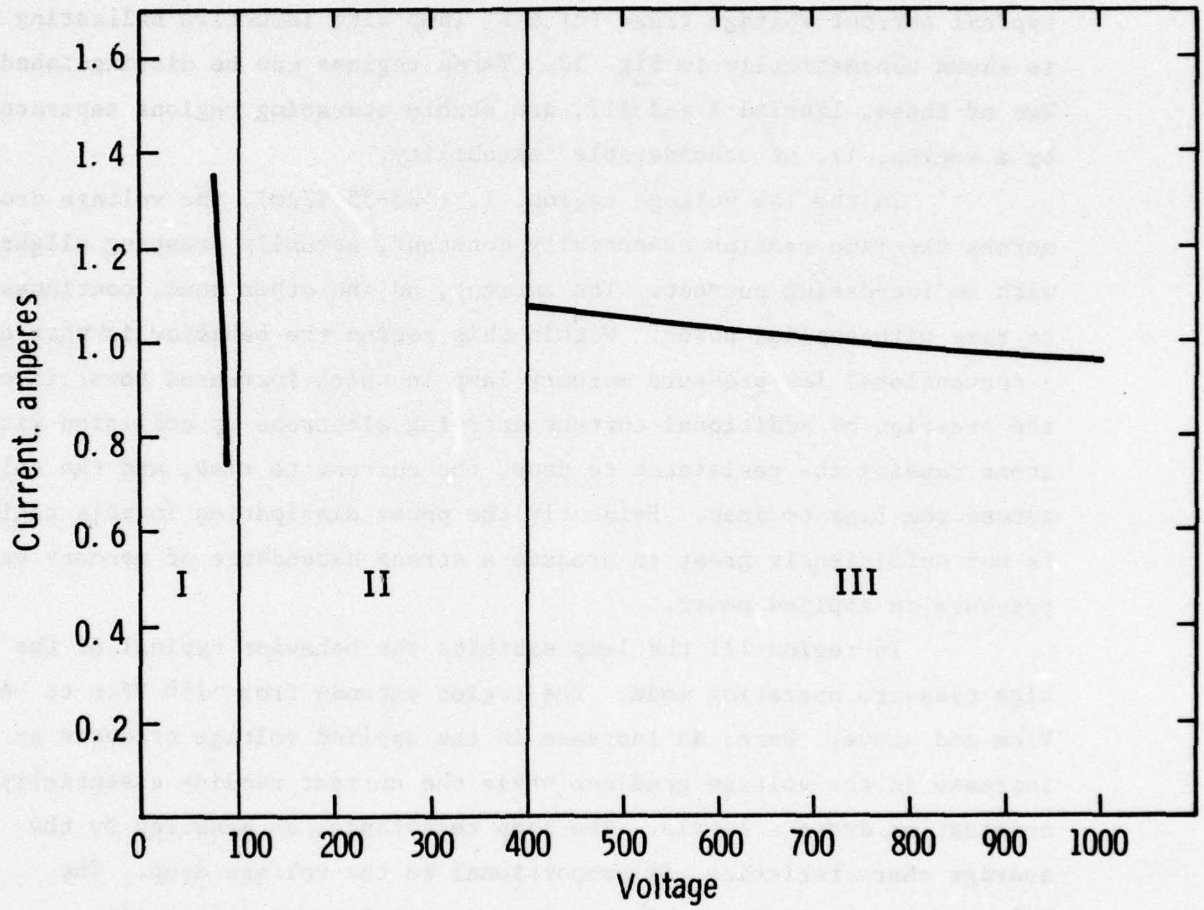

Fig. 12. Current-voltage characteristics of AH-6 lamps.

Within region II, the lamp acts as if it cannot decide between constant voltage mode or the constant current mode. The lamp is generally quite unstable and more difficult to ballast. While not accessible with water-cooled lamps, operation within this region is possible to a limited extent by air cooling.

4.3.2 Pressure Variation with Simmer Power

Since the simmer power establishes the lamp pressure, and the lamp pressure determines the number density of mercury atoms in the vapor, and hence the maximum radiant energy, it was desirable to determine the pressure variation with simmer power.

The mercury pressure in a lamp is established by the coldest part of the lamp. For a given lamp diameter, the pressure, arc temperature, current and voltage gradient are interrelated.[8] Figure 13 shows the pressure, and the approximate number density as a function of voltage gradient for a 2 mm bore lamp at two values of currents as deduced from data in references 4 and 8. The curve for the current of 1.25 amperes was obtained from the emperical formula of deGroot[9] for a 2 mm bore high pressure lamp:

$$P(\text{atm}) = \frac{E-100}{3} \quad .$$

where E is the voltage gradient in volts/cm.

Once the pressure is established, the temperature of the coldest part of the lamp can be estimated from published tables of vapor pressure data.[10] At 33 atm, which corresponds to operation at half rated power, this temperature is close to 870 K. The arc temperature, on the other hand, is probably between 5500 and 6500 K.

4.3.3 Comparative Light Output Measurements

It is well known that to pump a dye laser short pulses ($\lesssim$1 μsec) with high peak powers are needed. Although the original work by Dal Pozzo referred to pulses 30 μsec long, we made an effort to shorten the pulses in order to obtain higher peak powers for the same input energy. The

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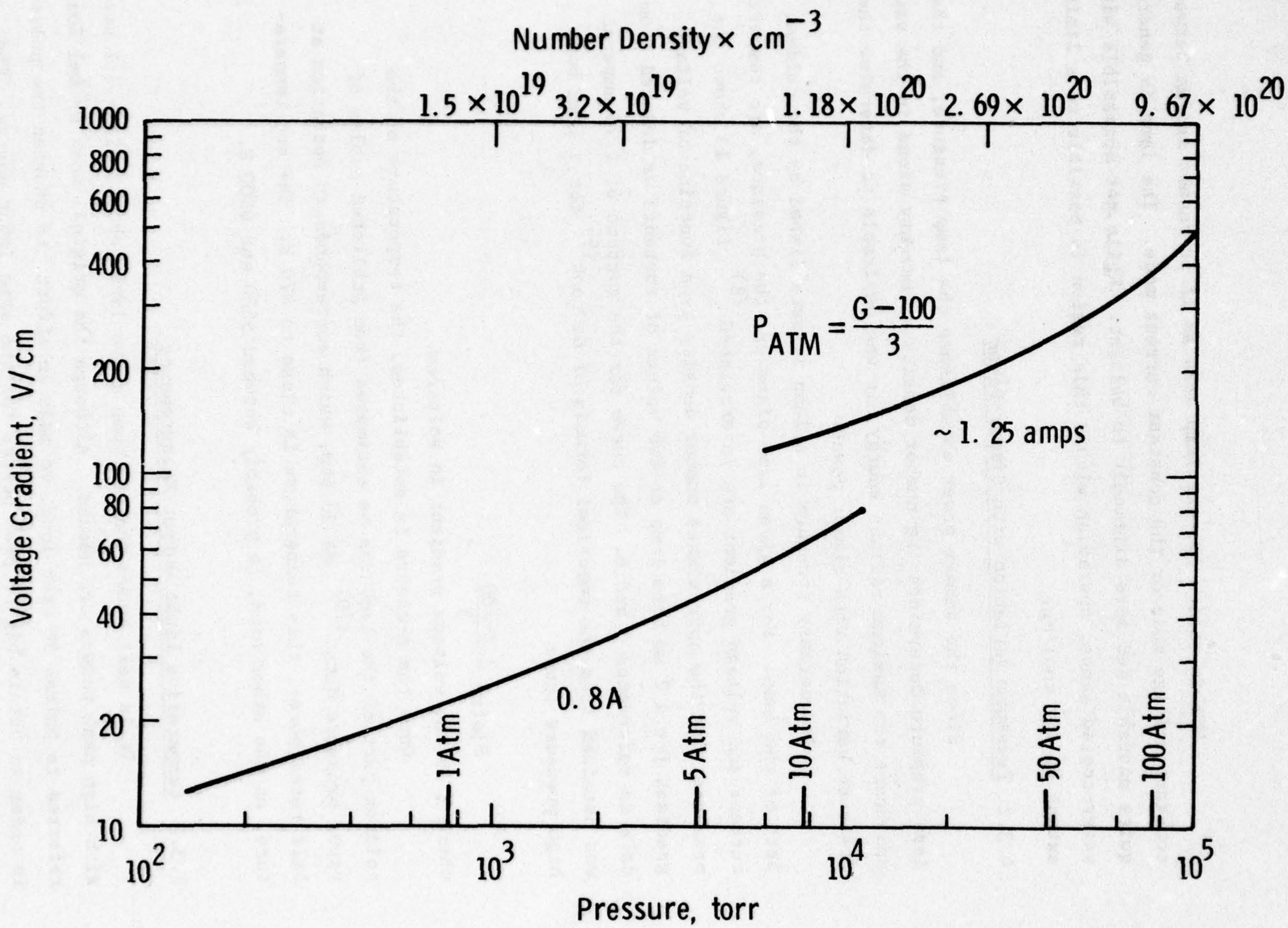


Fig. 13. Pressure vs voltage gradient for a 2 mm bore mercury arc lamp.

radiant pulse width and peak powers were measured with the photodetector filter combination used to simulate a blue dye excitation. A second motivation for these measurements was to attempt to lower the simmer power which, if operated in the manner of Dal Pozzo et al, would consume 1000 watts in a 2" lamp, and would contribute greatly to excessive heating of the dye and to an overall low electrical efficiency. Part of the effort to lower the simmer power involved operating the lamp air cooled.

Comparative light output measurements of peak pulse radiant intensities were made under various conditions of simmer operation. In general, the peak radiant power of the pulsed output as measured by the standard filter-detector combination increased almost linearly with input energy, for a given pulse width; for a given input energy, the peak radiant power also increased with a reduction in pulse width, as might be expected. Maximum peak radiant power was obtained operating aircooled, at a mercury pressure of about 1-3 atm, and a density of $\sim 1 \times 10^{19}$ cm^{-3}.

Figure 14 shows some comparative light measurements in arbitrary units, as a function of input energy. The electrical pulse width was ~ 5 μsec. Shown are results obtained with an AH6 mercury capillary lamp operated at high simmer power (50% of rated power) and relatively high pressure (~ 30 atm). Also shown is a similar lamp operated water cooled but in the very low pressure mode (~ 250 Torr). It is evident that the high pressure lamp radiates more strongly in the desired spectral region than does the low pressure lamp. Better results were obtained by operating the lamps air cooled at a pressure of 1-3 atm, although direct comparisons were not made because of differences in the test set ups.

In general, it is evident that a minimum vapor pressure is needed to provide suitably high radiant fluxes, although this pressure need not be as high as 30 atm.

Comparisons were also made between a mercury lamp operating air cooled at a pressure of ~ 3 atm, with 1 μsec pulses, and a commercial xenon lamp, ILC L-2589, operating air cooled with 1 μsec pulses. For these measurements, the xenon lamp was also operated on a simmer mode. An attempt to operate a water cooled mercury lamp at 30 atm with 1 μsec pulses resulted in destruction of the lamp.

Curve 695108-B

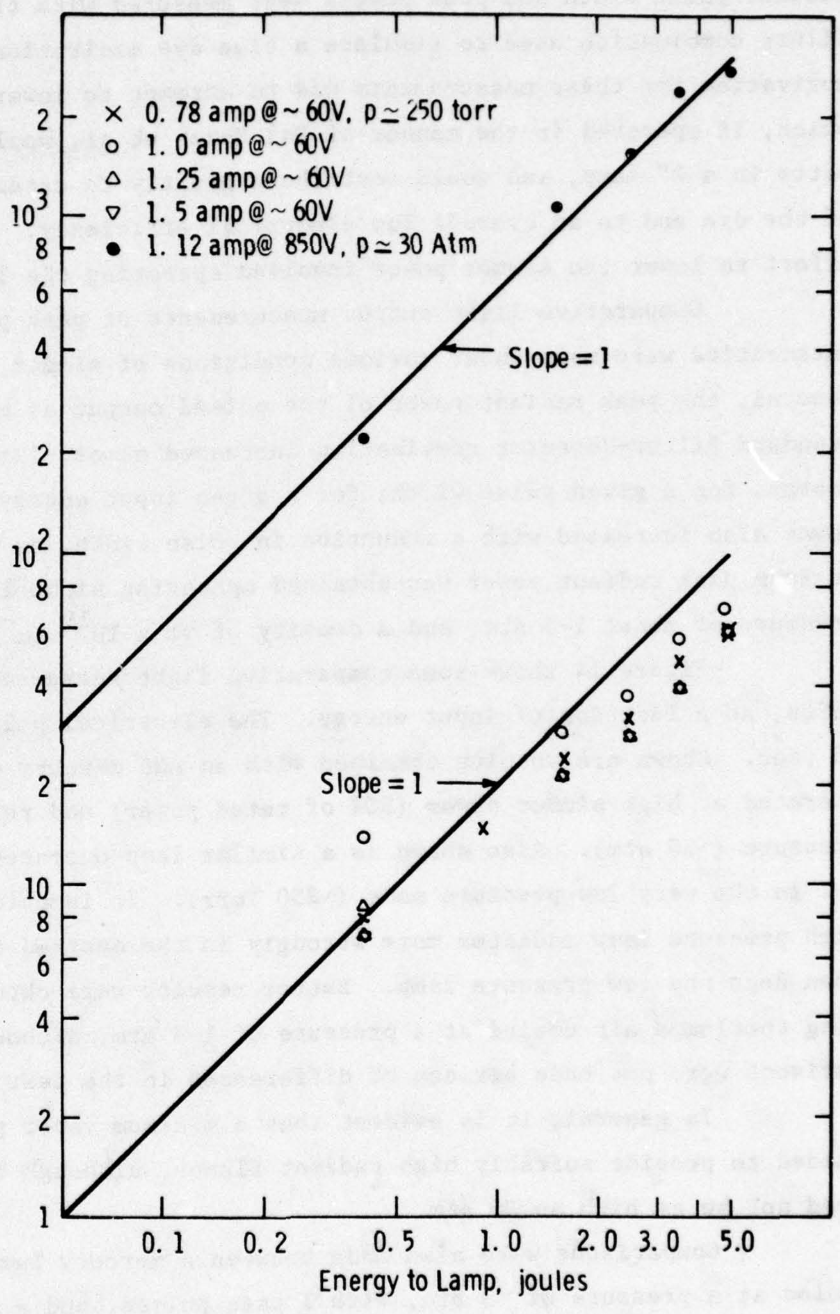


Fig. 14. Relative peak radiant power in the coumarin pump band region vs energy input of an AH-6 lamp at two operating pressures.

In addition, measurements at 30 atm were made with pulses approximately 50 μsec long, in order to simulate the conditions of references 1-3. Peak powers under these conditions were very low, and it is likely that the pulse shape differed considerably from that of Dal Pozzo et al.

Figure 15 shows typical light and current pulse shapes for xenon and mercury with 1 μsec pulses and mercury at 30 atm with a 50 μsec pulse.

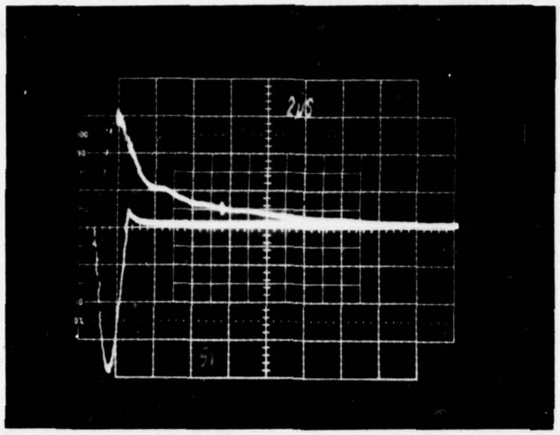
A further comparison between the air cooled mercury lamp and the air cooled xenon lamp are shown in Table I. The mercury lamp was operated with a simmer current of 125 ma and a voltage gradient of 40 V/cm. This corresponds to P ≃ 1 Atm. The xenon lamp was an ILC-L2589 operated with a 50 ma simmer current.

It is evident from Table I, that a mercury lamp, operated in the above manner is not very different from a similarly operated xenon lamp. This conclusion is further supported by the results of laser tests, described in Section 4.3.4 below.

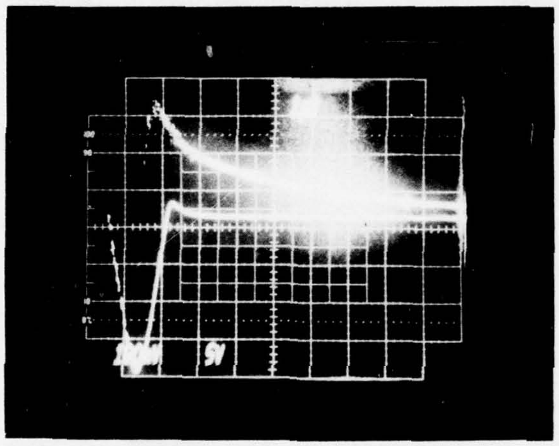
Some differences between mercury and xenon may be noted. With 1 μsec pulses, mercury was deposited on the walls of the capillary lamp, causing a steady reduction of lamp output with each pulse. This was attributed to two causes: 1) the fact that these were overdosed lamps with a large surplus of liquid mercury that was present at the electrodes and 2) the inability of the electrodes to easily provide the high peak currents. This caused electrode material, mainly mercury, to be sputtered on the inner walls of the lamp. Most of the lamp properties could be restored after one or several pulses by heating the lamp and forcing the mercury to redeposit near the electrodes. There remained, however, after each pulse, a small residue that could not be removed, and that grew worse with continued operation. This problem can probably be eliminated by using dosed lamps with the proper fill, and by larger electrodes.

4.3.4 Comparative Laser Tests

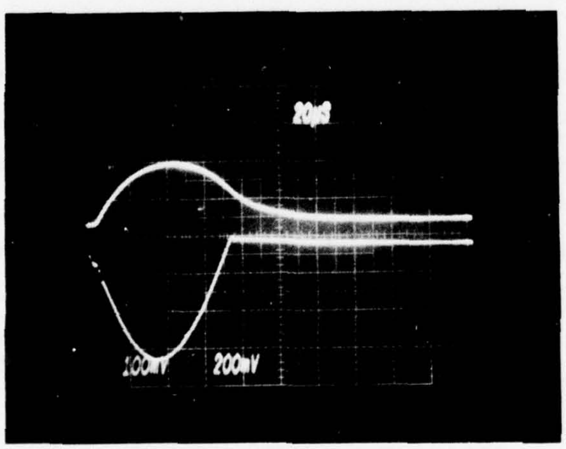
An initial assessment of the dye laser apparatus including both the spherical cavity and the pulsed power supply was made by comparing



a - Xenon lamp (ILC-L2589), air cooled, 10 joule, 1 μsec electrical pulse; upper trace, light pulse; lower trace, current pulse; horizontal scale, 2 μsec/div, vertical current scale, 1000 amp/div.



b - Mercury lamp, p $\sim$ 1-3 atm air cooled, simmer power 125 ma @ 200 V; conditions are same as in Fig. a, above except that horizontal scale is 1 μsec/div.



c - Mercury lamp, p $\sim$ 30 atm, water cooled, simmer power 1.25 amp @ 1000 V; 9 joules, 50 μsec electrical pulse; upper trace, light pulse; lower trace current pulse; 20 μsec/div; current scale, 40 amp/div.

Fig. 15. Current pulse and light pulse waveforms for xenon and mercury lamps with different simmer conditions.

Table I: Light Characteristics of Pulsed Xenon and Mercury Lamps

	Hg	Xe
Light Pulse		
rise time, nsec	∿ 200	∿ 500
decay time, μsec	∿ 2	∿ 1.5
pulse width, FWHM, μsec	1.5	2
relative peak detector signal, mv	∿ 300	∿ 320
Current Pulse		
pulse width FWHM, μsec	0.9	1.1
peak current, amperes	4200	4000
energy in discharge capacitor, joules	9.8	9.8

the threshold for Rh6G with that obtained with a Chromatix CMX-4 commerical flashlamp pumped dye laser. In order to make the comparison valid, a 4" long dye cell was used in the spherical cavity, this being the length of the Chromatix dye cell. Identical xenon filled flashlamps were used, both operated with simmer current. Also, the same dye module and dye solutions were used for both systems.

The results of the comparison of the two systems yielded a 4 joule threshold for the spherical cavity and a 6.5 joule threshold for the Chromatix CMX-4. These differences are very likely due to the different resonator reflectivities and not to the relative merits of the two systems. The comparison did serve to establish the adequacy of the spherical cavity for pumping dye lasers.

Subsequent tests were performed with a 2 inch long dye cell and 2 inch xenon flashlamp (ILC type L2589). A somewhat higher threshold was obtained due to the larger contribution from the end losses. (The end losses remained constant and independent of the length of the dye cell.) The 2 inch xenon lamp was used primarily for comparisons with the 2 inch pulsed mercury lamp. As in the comparative light output tests, the lamp was operated with a 50 ma dc simmer current. The pulse duration was 1.5 μsec.

Figure 16 shows the energy input vs. dye laser output using the xenon lamp to pump the dye. A slope efficiency of 3.2×10^{-4} was obtained.

Similar measurements were repeated with mercury lamps. Laser output was readily obtained operating air cooled with a 0.125 A simmer ($P \simeq 1$-1.5 atm) and 1.5 μsec pulses. The threshold, as determined by observing the lowest energy at which fairly consistant lasing took place was essentially the same for the mercury as for the xenon lamp. A fair measurement of the slope efficiency could not be obtained, however, because of the accumulation of mercury on the lamp wall, as discussed in Section 4.3.3.

Shown below are the thresholds obtained with mercury and xenon lamps in side-by-side comparison tests. The differences between the two thresholds are not considered significant. The threshold for the mercury

Curve 695107-A

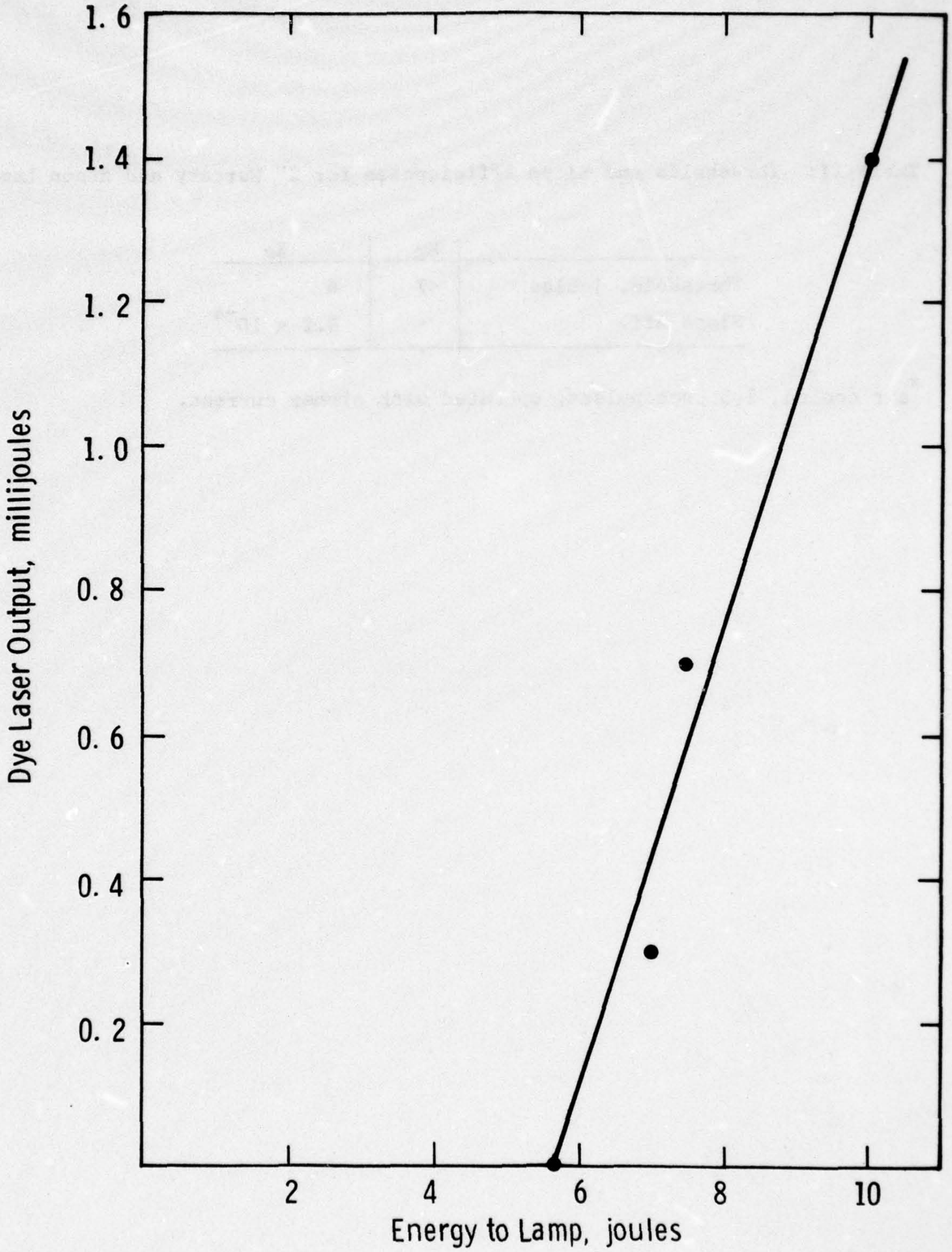


Fig. 16. Slope efficiency of xenon pumped dye laser.

Table II: Thresholds and Slope Efficiencies for 2" Mercury and Xenon Lamps*

	Hg	Xe
Threshold, joules	<7	6
Slope Eff.	-	3.2×10^{-4}

*air cooled, 1.5 μsec pulses, operated with simmer current.

lamp is probably lower than shown, because lamp output diminished even during the alignment necessary to obtain threshold values.

Additional measurements were performed with mercury lamps operated at 1000 watts simmer power and $\sim$30 atm pressure. Operating under these conditions created some additional problems because of the enormous amount of simmer power that needed to be dissipated. Much of this power is in the form of radiation that was focussed directly on the dye cell. The lamp operated in a much more stable fashion, however, and after long periods of time, there were no visible signs of deterioration or mercury accumulation on the lamp wall. Unfortunately, we were unable to obtain laser action. Use of long pulses ($\sim$50 μsec) in an effort to reproduce the results in references 1-3 were also unsuccessful. The pulse shapes, particularly the rise time, were probably quite different from those of Dal Pozzo et al.

4.4 Mercury Additive Lamps

4.4.1 Spectral Overlap Calculations

The most promising additives for the pump lamp were selected on the basis of 1) the spectral overlap between the lamp radiation and dye absorption, 2) the possibility of attaining adequate peak pump power radiated in the appropriate spectral band and 3) the possibility of obtaining adequate additive vapor densities, and pressures at attainable reservoir temperatures in quartz arc lamps.

The absorption spectra of four of the dyes mentioned in Section 4.2 were used as a basis for selecting the additives. These dyes were: 1) Coumarin 450 in 50-50 water and ethanol; 2) coumarin 466 in methanol; 3) coumarin 481 in dioxane; and 4) coumarin 485 in dioxane. Spectral absorption functions $F_i(\lambda)$ were determined by normalizing these absorption curves to a maximum absorption of unity for each dye. In addition, a "Superdye" with a spectral absorption of 1.0 for $365 \leq \lambda \leq 425$ nm and zero elsewhere was considered. An existing computer program was modified to give spectral overlap calculations for each of these five functions over a temperature range of 3000-10,000°K for seventy elements for which NBS spectral line intensity data were available.

For each of these elements three quantities were calculated for the neutral and singly-ionized species, neglecting self-absorption. These were: 1) the total power radiated in watts/cc, $P_R(T) = \Sigma I(\lambda,T)$, 2) the total power absorbed by the dye $P_A{}^i(T) = \Sigma I(\lambda,T)F_i(\lambda)$, and 3) the ratio of the power absorbed to that radiated

$$R(T) = \frac{P_A{}^i(T)}{P_R(T)} = \frac{\Sigma I(\lambda,T)F_i(\lambda)}{\Sigma I(\lambda,T)} .$$

Partition functions were calculated for the neutral and singly ionized species and the same three quantities were calculated for equilibrium neutral and singly-ionized species in thermodynamic equilibrium with electron densities of 10^{15}, 10^{16}, 10^{17}, and 10^{18} per cc.

Lamp power considerations place limits on the lamp size and the additive pressures. For the purposes of the present calculations, we assumed a requirement of 10 joules/pulse and a pulse repetition rate of 100 pps. This corresponds to an average power dissipation of 1000 watts. A power dissipation of this magnitude is compatible with a 10 cm long capillary lamp with a 2 mm bore.

A second requirement involves peak power. For a maximum pulse width of 10 μsec, the minimum peak power required is 10^6 watts. This must be produced in a radiating volume of 10 cm x 0.005 cm^2, where the effective radius of the radiating plasma is taken to be ≈0.4 of the actual radius. Thus, a peak power density from the lamp of $\sim 2 \times 10^7$ watts/cm^3 is required to meet the above criteria. The requirement of this fairly high power density can be relaxed somewhat by assuming a slightly larger lamp diameter.

Table III summarizes the results of our computer calculations. The ratio of the power absorbed by the dye to the total radiated power, R(T), defined above, is used for initial screening of potential additives. This ratio is shown in the second column. The potential additives are ranked according to values R(T) $\gtrsim$ 0.42. The <u>total</u> power absorbed by a dye is given by R(T) x $P_R(T)$ where $P_R(T)$ is the total power radiated at a temperature T. Values of this product are shown in the third column. Values of R(T) and $P_R(T)$ were obtained for an electron density of 10^{18} cm^{-3}

and a ratio of neutral to singly ionized species of roughly unity. To obtain these conditions, an additive pressure near 1 atm is needed. The fourth column shows the effective arc temperature, T, for the corresponding values R(T) and $P_R(T)$. The last column shows the approximate reservoir temperature that is needed to achieve a partial pressure of 1 atm for each species. Except for Hg, only halogens of the metals have high enough vapor pressures to be compatible with lamp reservoir temperatures. Thus, the values given in the last column are for the iodides of the metals shown.

The results of Table III need to be interpreted with caution and to be used only as a guide to potential pump lamp additives. They are based on atomic and ionic line radiators assumed to be in or at least near local thermal equilibrium. No self-absorption has been taken into account. For the rare earth and transition elements this is probably not a serious shortcoming. However, the radiation from Tl and Hg will be greatly reduced by self-absorption, i.e., by at least a factor of 3 to 8.

No radiation from molecular species has been taken into account. When mercury plus a metal iodide additive is used, some radiation from the HgI B → X transmission in the blue will be beneficial.

The additives listed in Table III can be classified into two groups: those with few lines in the region of interest, hence subject to considerable self-absorption, and those with many lines, and therefore less subject to self-absorption. In the first category are mercury and thallium (as TlI). In the second category, the species that had good radiant power, and low enough plasma temperatures and reservoir temperatures to be within reachwere Th, Sc and Pd.

4.4.2 Additive Lamps Measurements

A number of lamps were constructed and filled with mixtures based on the calculations described in the preceding section. All of the lamps contained some mercury in addition to the additive. Three such lamps are shown in Fig. 17 (b,c and d). Figure 17(a) shows a normal 10 cm x 2 mm high pressure AH-6 mercury lamp. Most of the lamps were

Table III: Ranking of Additives According to Their Calculated Effectiveness

Element	R(T)	$R(T) \times P_R(T)$ watts·cm^{-3}	T plasma °K	T reservoir* °K
Ho	0.73	5.07×10^6	8000	(1570)+
Dy	0.73	4.99 x "	"	(1590)
Er	0.68	5.21 x "	"	(1550)
Tb	0.65	2.89 x "	"	(1600)
U	0.65	2.29 x "	"	?
Gd	0.61	4.92 x "	"	(1610)
Th	0.55	2.29 x "	"	1110
Sc	0.54	3.30 x "	9000	1182
Yt	0.52	2.04 x "	"	1580
Tm	0.47	4.42 x "	8000	(1530)
Zr	0.46	3.32 x "	10,000	(1300)
Pd	0.43	7.91 x "	"	(1100)
Tl	0.43	1.42 x "	"	1096
Hg	0.42	2.76×10^8	"	630‡
Ti	0.42	3.38×10^6	9000	(1300)

* as metal iodides

\+ parentheses denote less certain values

‡ as metal

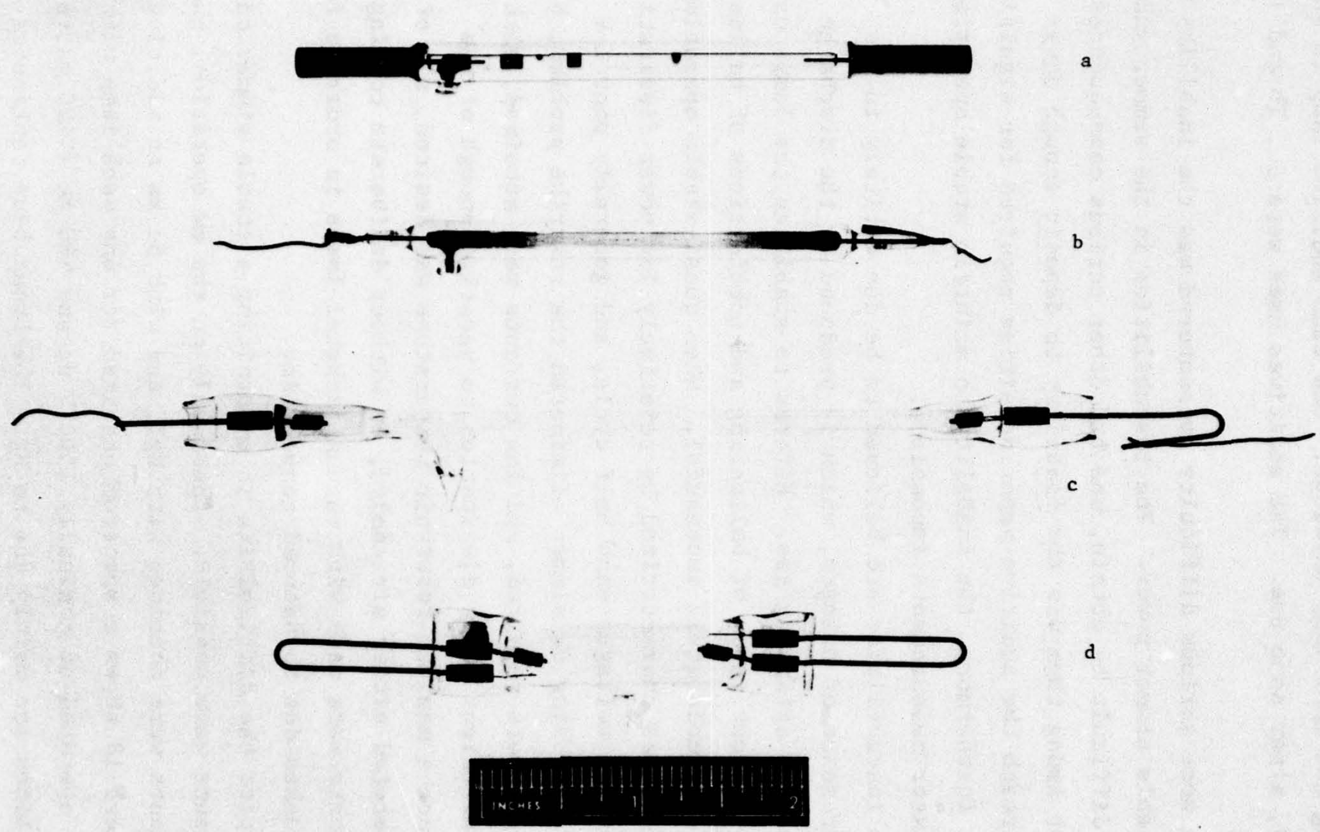


Fig. 17. Mercury and mercury plus additive lamps. From top down: a) AH-2B high pressure mercury lamp; b) AH6-2B lamp filled with Sc, I_2, and Hg, after use; c, and d, experimental lamps with two different bores.

made utilizing the AH-6 lamp envelope. One such additive lamp is shown in Fig. 17(b), after some use. The additives used were Sc, Th and I_2 (as HgI_2).

The most serious difficulty encountered was the inability to maintain a stable simmer power. The instabilities in the simmer made measurements difficult to obtain, and had other serious consequences. Most important among them was the inability to deposit enough power in the lamps to reach the additive vapor densities required for significant enhancement. Furthermore, the inability to maintain stable operation made pulsed laser measurements impossible.

The instabilities are believed to be due entirely to the unavoidable presence of halogen, which is produced by the discharge and is an electron attaching gas. Efforts to stabilize the lamps using ac simmer and various types of ballasting and combinations of ballasts proved to be only moderately successful. When quasi-stable operation was achieved, it was characterized by relatively low power dissipation, very high restrike voltages each half cycle, and generally poor I-V characteristics. Using dc simmer eliminated the restrike problem, but high dc voltages were required, and low currents were attained, with resulting insufficient power dissipation to vaporize enough of the additive. Since a maximum reservoir temperature was desired, all of the lamps were operated either air cooled, or without deliberate cooling. Some measurements were made with vacuum jacketed lamps in order to further reduce heat losses due to natural convection.

Despite the difficulties in maintaining a stable simmer current, some measurements were possible. Based only on the cw operation, the best enhancements were obtained with HgI_2 and with Sc as an additive.

Figure 18 shows a spectrum obtained for one such lamp with an AH-6 envelope operated at nominally 1500 V dc and $\sim$80 ma ($\sim$120 watts) simmer. The lines are mainly due to Hg. The broad blue continuous band extending from 350 nm to 450 nm is the molecular band of HgI. Weak Sc emission is seen at 390.7, 391.1 nm, and at 402.3 nm.

Figure 19 shows a similar composition in an 8 mm bore envelope. The larger bore resulted in a higher current, $\sim$0.8 amp, and lower

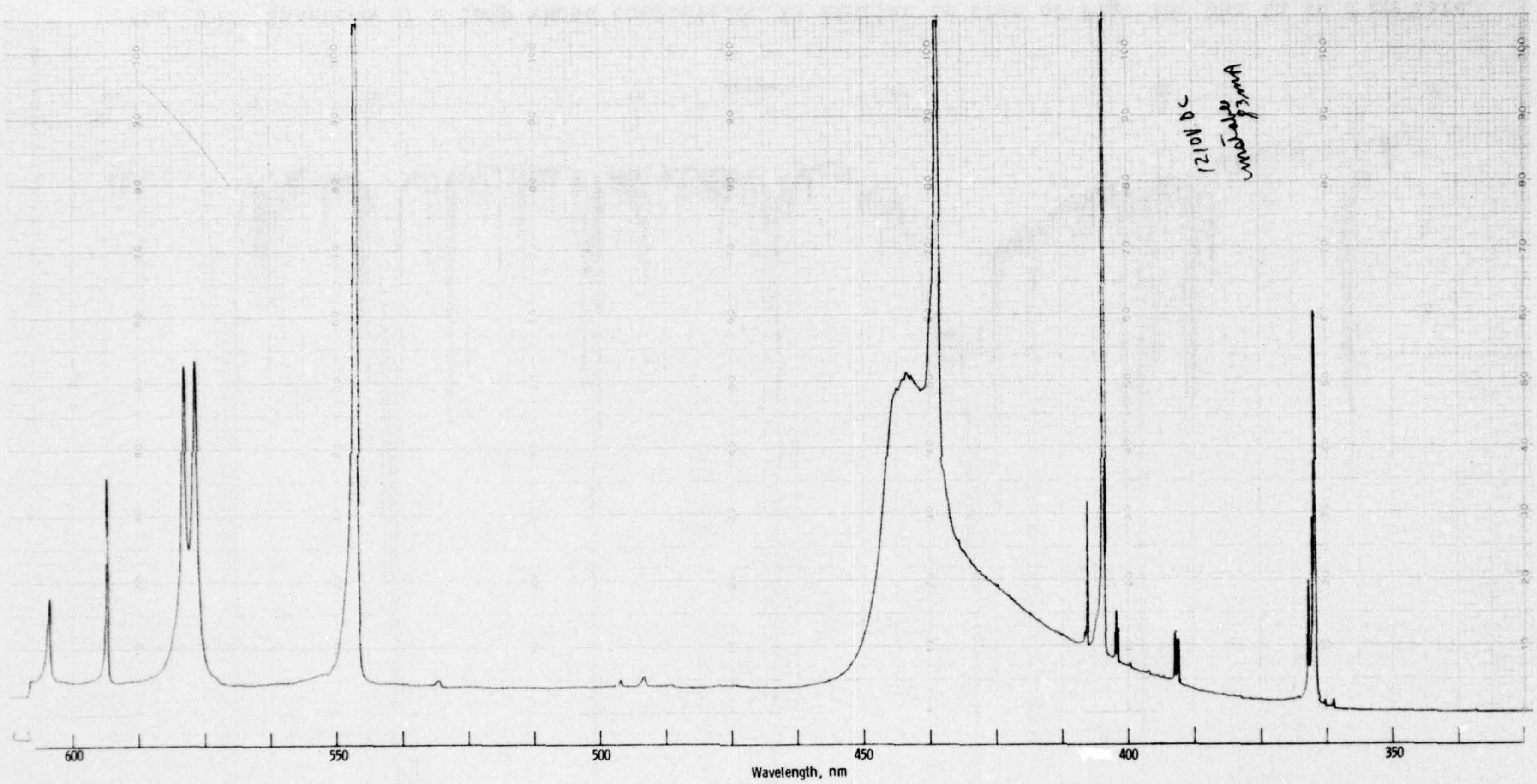


Fig. 18. Spectrum of a lamp containing Hg, Sc and I_2 in an AH-6 envelope, using dc simmer, operating in air.

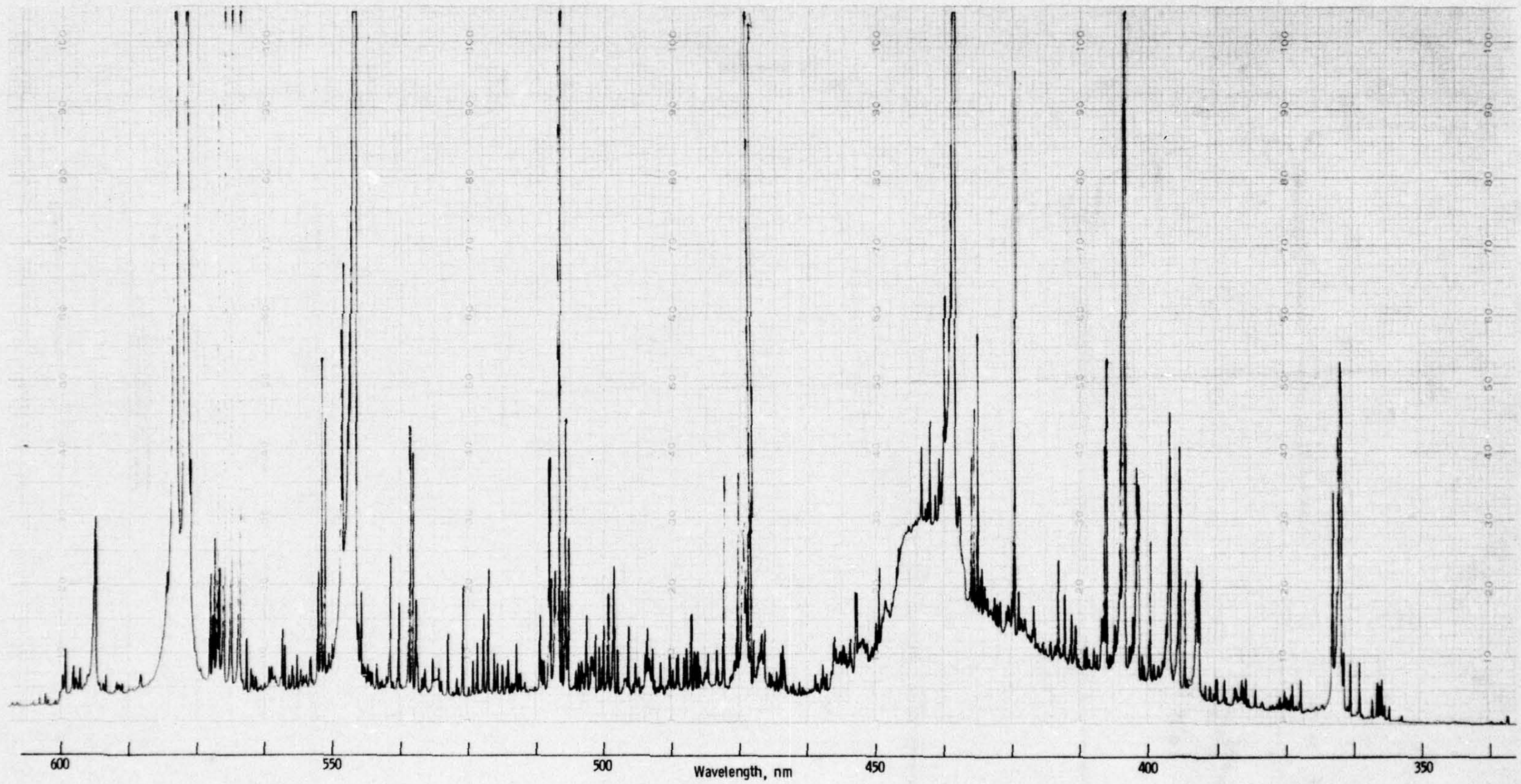


Fig. 19. Spectrum of a lamp whose composition is similar to that of Fig. 18, but in an 8 mm bore envelope.

voltage, ~250 V dc, for a dissipation of 200 watts. This lamp gave a much greater contribution to the spectrum from Sc, as evidenced by the very large number of lines seen in the spectrum. (Compare with Fig. 18.) The lamps were not able to sustain this power loading for any great length of time, and this lamp exploded shortly after the spectrum was obtained.

Of the additives tried, HgI_2 appeared to be the most promising, and one that could be operated at reasonable temperatures. Accordingly, some comparison tests, similar to those described in 4.3.3, were made *between AH-6 lamps containing mercury, and those containing* HgI_2. In these tests, no improvement was seen due to the added iodine, although the compositions may have been far from optimum.

5. DISCUSSION

The conclusions that were reached after testing lamps containing mercury alone and with additives are as follows:

1. While mercury filled flashlamps can be used to pump dye lasers, at the present time they do not appear to offer significant advantages over conventional xenon flashlamps.

2. The effectiveness of mercury lamps depends very much on how they are operated. The most effective pressure range appears to be about 1-3 atm. Low pressure lamps ($p \lesssim 100$ torr) cannot supply sufficient radiant power density. Operating at high mercury pressures ($p \sim 30$ atm) we were unable to pump a laser dye, unlike Dal Pozzo et al,[1-3] although our conditions were sufficiently different as to make comparisons difficult. Nevertheless, the high simmer power required to operate in the high pressure range imposes a very high heat load on the dye, exposes the dye to excessive radiation, and seriously affects any gains in efficiency that might be obtained because of better spectral properties. Operating at high energies ($\gtrsim 10$ joules per pulse) and high repetition rates ($\gtrsim 100$ pps) could greatly reduce or eliminate the simmer power, but we cannot say whether such operation is possible with capillary lamps, or desirable. It is likely that in the $\sim$1-3 atm pressure range some improvement in the performance of mercury lamps, at least as far as stability and consistent performance are concerned, can be achieved through the use of dosed lamps, fitted with heavier electrodes which are better capable of handling the high peak currents.

3. The similarity in behavior between the medium pressure air cooled mercury lamp and a xenon lamp of roughly comparable pressure suggests that both lamps are radiating essentially as black bodies. (This point is discussed further, below.)

4. With regard to the additive lamps, the limited spectra obtained were consistent with the computer predictions. The difficulties

arose because of an inability to maintain stable operation, and a failure to reach temperatures high enough to provide suitable vapor densities. Although stable operation without simmer power may be possible using very short pulses ($\lesssim$ 100 nsec, i.e., shorter than the time required to establish an arc discharge) or by means of other additives, such an investigation would have required a greater effort than was possible on this project.

6. DISCUSSION

The observation that at the pressure where the mercury lamp ($p \approx 1$ atm) was able to pump a laser mercury and xenon behaved alike suggests that both lamps radiated like black bodies over a significant portion of their spectrum. That such is the case for xenon is generally known. It is known, for example, that at high current densities, the visible through infrared spectrum of a xenon lamp resembles that of a black body of $T \approx 9{,}500$ K.(10) Figure 20 shows the spectrum of a double pulsed xenon lamp at a current density of 43,000 amp/cm^2 obtained some time ago in these laboratories. The spectrum is well matched by a 9500 K black body from 300-600 nm.

The situation might be expected to be somewhat different for the case of mercury, since mercury is known to emit a considerable amount of its radiation as molecular radiation produced by mercury dimers. However, calculations using the results of Zollweg, Liebermann and Burnham(6) indicates that at temperatures of 6000 K and above and for very high peak current densities, of the magnitude we have measured, recombination radiation and bremsstrahlung radiation are more intense than molecular radiation in the range from 350 to 700 nm, and that the emissivity and spectral radiance in this range are essentially that of a black body. Clearly as the spectrum of a lamp approaches that of a black body, the advantages of a selective radiator are diminished.

One may view the trend towards black body radiation as coming about through the action of collisions. Both high pressures and high current densities such as occur during short pulses and high peak powers, drive the lamp radiation in the direction of that of a black body. Pulsed laser dyes are presently limited to laser pulse durations of not much longer than 1 μsec. This means that a flashlamp whose pulse duration exceeds 1 μsec becomes progressively less efficient with increasing pulse

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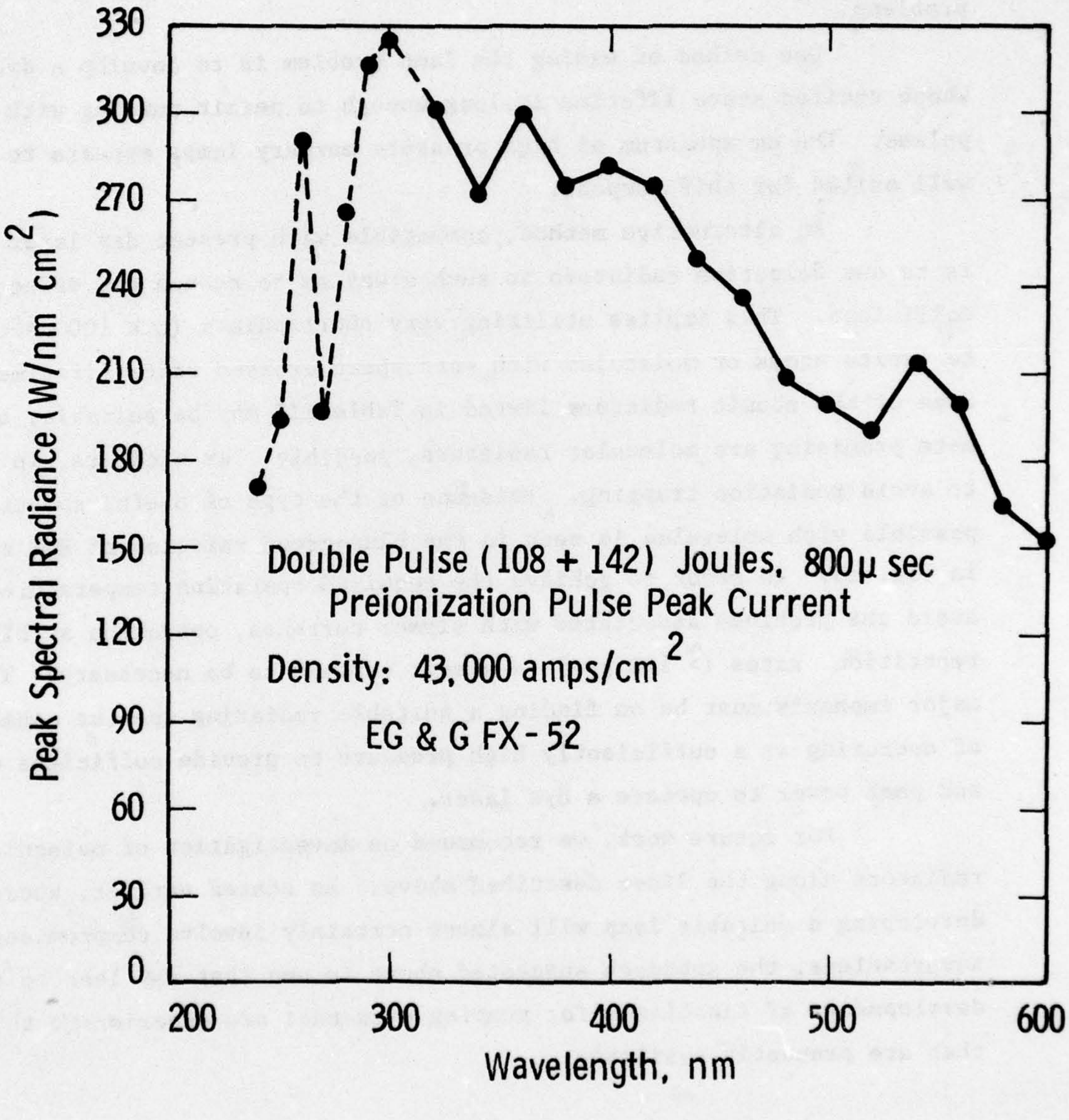

Fig. 20. Spectrum of a xenon lamp operated double-pulsed at high current density.

length. The short excited-state lifetimes of most dyes also requires of the flashlamp a high peak radiance. Both requirements are therefore in opposition to the conditions that favor selective spectral radiators. Any successful spectral radiator must result from either a compromise among the operating conditions or from a circumvention of some of the problems.

One method of easing the lamp problem is to develop a dye whose excited state lifetime is long enough to permit pumping with long pulses. The cw spectrum of high pressure mercury lamps appears to be well suited for this purpose.

An alternative method, compatible with present day laser dyes, is to use selective radiators in such a way as to reduce the effect of collisions. This implies utilizing very short pulses ($t < 100$ nsec) to excite atoms or molecules with very short excited state lifetimes. Some of the atomic radiators listed in Table III may be suitable, but more promising are molecular radiators, possibly as excimers, in order to avoid radiation trapping. Evidence of the type of useful spectra possible with molecules is seen in the blue-green emission of HgI shown in Fig. 18. In order to achieve the required operating temperatures and avoid the problems associated with simmer currents, operation at high repetition rates ($\gtrsim 100$ pps) is almost certain to be necessary. The major emphasis must be on finding a suitable radiating species capable of operating at a sufficiently high pressure to provide sufficient energy and peak power to operate a dye laser.

For future work, we recommend an investigation of molecular radiators along the lines described above. As stated earlier, success in developing a suitable lamp will almost certainly involve compromises. Nevertheless, the approach suggested above is one that may lead to the developments of flashlamps for pumping dyes that are superior to those that are presently available.

7. ACKNOWLEDGEMENTS

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